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Supporting Information

Electrochemical single-step *N*-acylation and *S*-cyclization synthesis of thiazolimide via radical process

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1. Materials and instrumentation

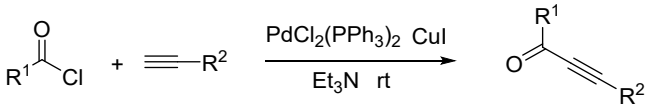
All starting materials and reagents were purchased from commercial sources and used as received unless otherwise mentioned. ^{1}H NMR, ^{13}C NMR and ^{19}F NMR spectras were recorded respectively on 400 MHz, 101 MHz and 376 MHz JEOL ECS-400 MHz spectrometer. Chemical shift values are given in ppm (parts per million) with tetramethylsilane (TMS) as an internal standard. The peak patterns are reported as follows: s, singlet; d, doublet; t, triplet; q, quartet; quint, quintet; m, multiplet; dd, doublet of doublets; dt, doublet of triplets; td, triplet of doublets. X-ray diffraction data were recorded on Bruker SMART APEX II CCD diffractometer.

2. Experimental procedure

2.1 General procedure for the synthesis of alkynones[1]

To a solution of alkyne (10.0 mmol) in anhydrous Et_3N (8.0 mL), was added with $PdCl_2(PPh_3)_2$ (10.0 mg, 14.3 μmol, 0.9 mol%) and CuI (11.0 mg, 58.0 μmol, 3.0 mol%). The reaction was left to stir for 5 min at RT. Then the acid chloride (10.0 mmol) which diluted in THF (1.0 mL) to the mixture dropwise under a N_2 atmosphere. The reaction mixture was then stirred for 5 h at room temperature and diluted with dichloromethane (25.0 mL) and washed with water (30.0 mL). The aqueous layer was then extracted with dichloromethane (20.0 mL) three times and all organics combined dried over anhydrous Na_2SO_4. Solvents were then evaporated under reduced pressure, and the residue was chromatographed on silica gel (petroleum ether/ethyl acetate =30/1) to give the pale yellow product.

Scheme S1 General procedure for the synthesis of alkynones

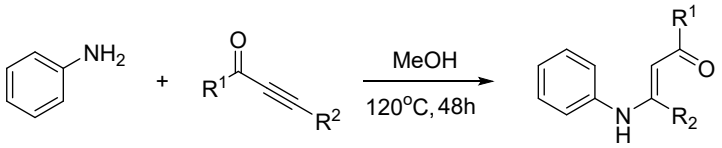


2.2 General procedure for the synthesis of enaminones[2]

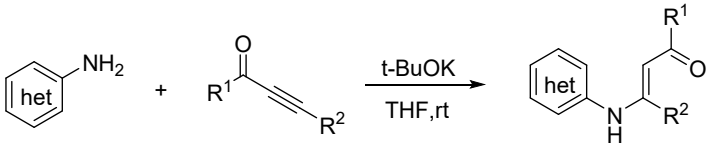
N-phenyl-substituted enaminones were general obtained by condensation of arylamine with alkynones in dry MeOH. To an over-dried schlenk tube, 1,3-diphenasylpropyl-2-yn-1-one (824.0 mg, 4.0 mmol), aniline (373.0 mg, 4.0 mmol) and anhydrous MeOH (1.5 mL) was added. Then sealed the reaction tube and stirred at 120 °C for 48 h until the reaction of the starting materials were completely disappeared. The reaction mixture was then cooled to room temperature and the residual solvent was

evaporated. Finally, the crude recrystallized with 95%-alcohol/petroleum ether to afford 1.37 g of the product as yellow crystals in 91% yield. The rest of the enaminones were synthesized with the same method and purified by recrystallization and column chromatography (petroleum ether /dichloromethane).

Scheme S2 General procedure for the synthesis of enaminones

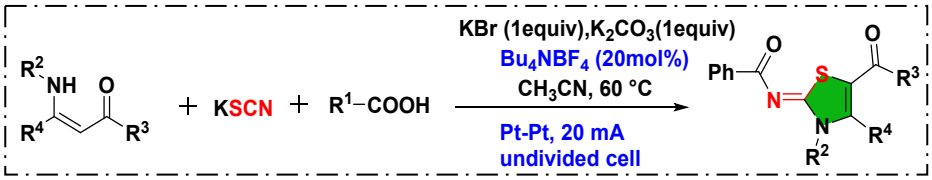


N-heterocyclic substituted enaminones were obtained by condensation of heterocyclic arylamine with alkynones in the presence of t-BuOK. Steps are as follows: To a solution of anhydrous THF (8.0 mL), heterocyclic arylamine (4.0mmol) and 1,3-diphenasylpropyl-2-yn-1-one (824.0 mg, 4.0mmol) were added. After substrates were thoroughly mixed, t-BuOK (897.0 mg, 8.0 mmol) was dropwise added to the mixture in fractions. After 1h, the reaction mixture was quenched by adding 25.0 mL distilled water, extracted three times with dichloromethane (25.0 mL), then the product was separated by concentration and column chromatography.



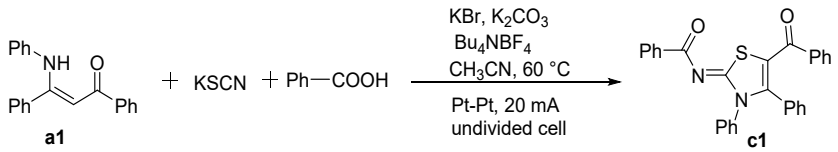
2.3 Synthesis of thiazolimide

In a 10 mL glassware charged with benzoic acid (0.5 mmol), enaminone (0.5 mmol), KSCN (0.6 mmol), accelerant (0.5 mmol), Electrolyte (20.0 mol %), K_2CO_3 (0.5 mmol) in CH_3CN (3.0 mL) were stirred at 60°C for 24h. The bottle was equipped with Pt wire electrode as the anode and cathode. The electrochemical workstation was used for 20 mA current catalysis. This electro-catalyst reaction progress was monitored by TLC, upon completion, the mixture was then extracted with dichloromethane (30.0 mL) three times and washed with saturated salt water. After evaporation of the solvent, the target crude product was obtained and purified column chromatography silica (petroleum ether/ethyl acetate=5/1) to afford the target product (**c1**) as pale yellow solid in 87% yield. The preparation of other corresponding products is similar to that of **c1**.

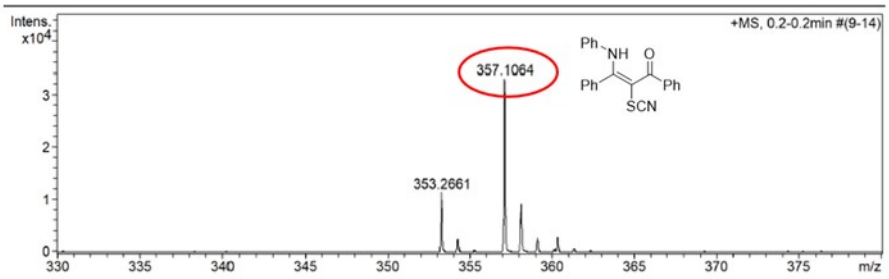


2.4 General procedure for the scaled-up experiment

In 10 ml glassware equipped with enaminone (1.0 mmol), benzoic acid (1.0 mmol), KSCN (1.2 mmol), accelerant (1.0 mmol), Electrolyte (0.2 mmol), K_2CO_3 (1.0 mmol) and a stir bar. The bottle was equipped with Pt wire electrode as the anode and cathode. Then 5 mL CH_3CN was added. The reaction mixture was stirred and electrolyzed at a constant current of 20 mA under 60°C for 24h. After the reaction is completed, the mixture was extracted with dichloromethane and washed with saturated salt water, the residue was purified by silica gel column chromatography to obtain the corresponding product (c1) as pale yellow solid in 83% yield.

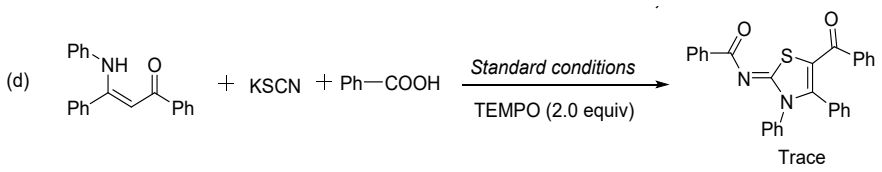


2.5 Mass spectrum of a1



2.6 General procedure of radical trapping experiment

In a 10 mL glassware, the 2, 2, 6, 6-tetramethylpiperidin-1-oxyl (2.0 mmol), enaminone (0.5 mmol), KSCN (0.6 mmol), benzoic acid (0.5 mmol), accelerant (0.5 mmol), Electrolyte (20.0 mol %), K_2CO_3 (0.5 mmol) and 3.0 ml of CH_3CN were placed. The mixture was stirred at 60°C for 24h. Then, the reaction mixture was stirred under 20 mA current catalysis. The reaction progress was monitored by TLC. After the reaction was completed, TLC showed that the reaction was suppressed.



3. Crystallographic details

The crystal of **c3**, **c8**, **c11** and **c34** were evaporated and cultured from ethyl acetate and *N*-methylpyrrolidone. For the single crystal analysis of these compounds, white block crystals were taken directly from the mother liquor, and transferred onto a loop. X-ray diffraction analyses were operated on a supernova diffractometer with a Multilayers mirror Ga-Kα radiation (λ = 1.35 Å) by using a ω scan mode at 150.0 K, 180.0 K, 180.0 K, 180.0 K, 100.0 K, respectively. Structures were analyzed by direct phase determination and refined by full-matrix least-squares method on F^2 with anisotropic thermal parameters for all non-hydrogen atoms (SHELXL-97)[3]. All non-hydrogen atoms were subjected to anisotropic refinement. Hydrogen atoms were placed in calculated positions and allowed to ride in their respective parent atoms. The thermal ellipsoid plots of crystal structures were recorded in Figure S1, Figure S2, Figure S3 and Figure S4, respectively. Further details of structural analyses of **c3**, **c8**, **c11** and **c34** were recorded in Table S1-S4. CCDC 2224992 (compound **c3**), CCDC 2224990 (compound **c8**), CCDC 2224996 (compound **c11**) and CCDC 2224993 (compound c**34**) contain the supplementary crystallographic data for this paper.

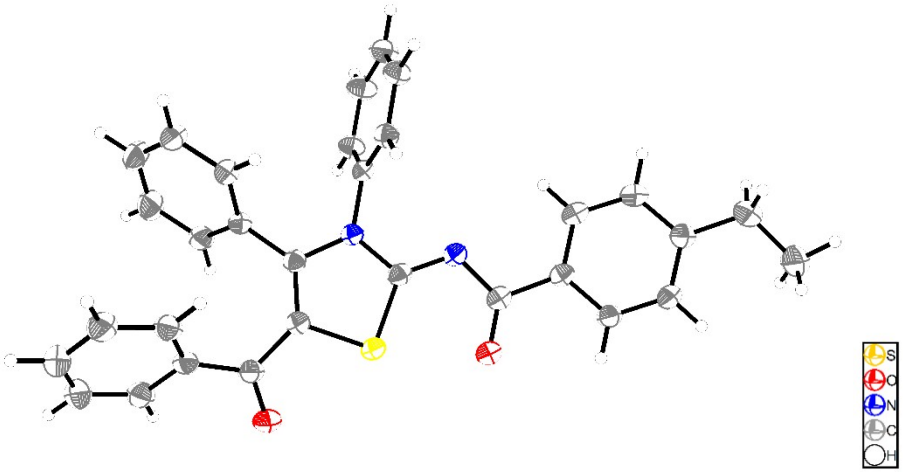


Figure S1 X-ray crystal structure of **c3** (CCDC number: 2224992)

Table S1 Summary of X-ray crystallographic data for **c3**

formula	$C_{31}H_{24}N_2O_2S$
formula weight	488.16
temperature (K)	180.0
crystemtal system	triclinic
Volume (Å3)	1266.2(7)
Space group	P-1
a, Å	9.279(3)
b, Å	11.406(2)
c, Å	13.046(5)
α, deg	80.40(2)°
β,deg	80.80(3)°
γ,deg	69.338(16)°
μ（mm-1）	0.954
Z	2
Density (mg.cm-3)	1.289
F(000)	515
Reflections collected/unique	18580/4868
Rint	0.0489
Goodness-of-fit on F2	1.050
R indices (all data)	0.0460/0.0958
Final R indices[I>2σ(I)]	0.0395/0.1040

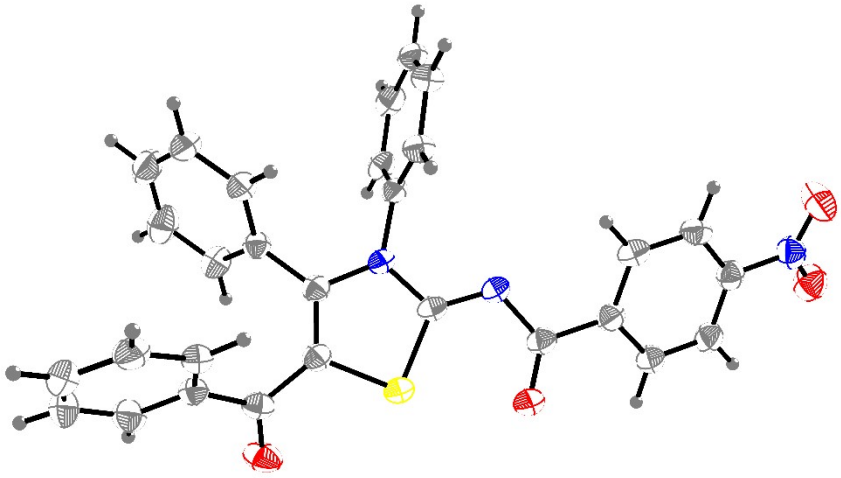


Figure S2 X-ray crystal structure of **c8** (CCDC number: 2224990)

Table S2 Summary of X-ray crystallographic data for **c8**

formula	$C_{29}H_{19}N_3O_4S$
formula weight	505.11
temperature (K)	180.0
Volume (Å3)	1212.9(4)
Space group	P-1
a, Å	9.5210(11)
b, Å	12.027(3)
c, Å	12.459(2)
α, deg	63.128(14)°
β,deg	82.674(12)°
γ,deg	72.395(15)°
μ（mm^{-1}）	1.038
Z	1
Density ($mg.cm^{-3}$)	1.389
F(000)	526
Reflections collected/unique	24305/4577
R_{int}	0.0368
Goodness-of-fit on F^2	0.990
R indices (all data)	0.0361/0.0990
Final R indices[I>2σ(I)]	0.0358/0.0988

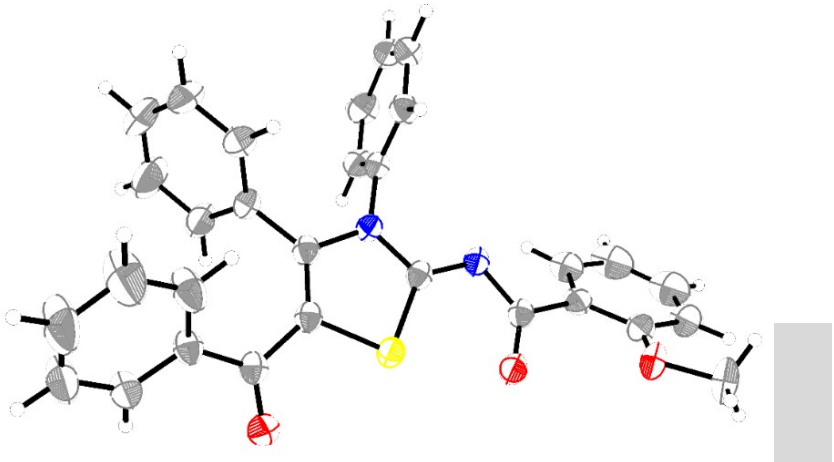


Figure S3 X-ray crystal structure of **c11** (CCDC number: 2224996)

Table S3 Summary of X-ray crystallographic data for **c11**

formula	$C_{30}H_{22}N_2O_3S$
formula weight	490.14
temperature (K)	180.0
Volume (Å3)	2459.4(8)
Space group	P-1
a, Å	11.803(3)
b, Å	12.782(2)
c, Å	17.329(2)
α, deg	89.118(10)°
β,deg	81.911(12)°
γ,deg	71.924(15)°
μ（mm^{-1}）	1.136
Z	2
Density ($mg.cm^{-3}$)	1.351
R_{int}	0.0611
F(000)	1042
Reflections collected/unique	9876/2459
Goodness-of-fit on F^2	1.031
R indices (all data)	0.0427/0.0587
Final R indices[I>2σ(I)]	0.0406/0.0916

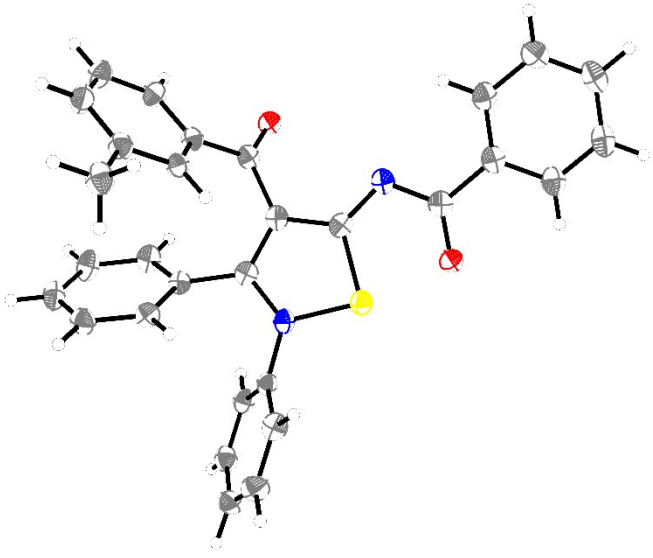
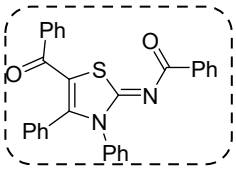


Figure S4 X-ray crystal structure of **c34** (CCDC number: 2224993)

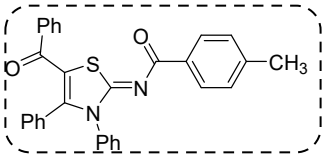
Table S4 Summary of X-ray crystallographic data for **c34**

formula	$C_{30}H_{22}N_2O_2S$
formula weight	474.14
temperature (K)	100.0
Volume (Å3)	1199.6(3)
Space group	P -1
a, Å	9.9070(13)
b, Å	11.5183(16)
c, Å	11.8511(10)
α, deg	108.421(10)°
β,deg	104.160(9)°
γ,deg	99.584(12)°
μ（mm^{-1}）	0.938
Z	2
Density ($mg.cm^{-3}$)	1.314
R_{int}	0.0512
F(000)	496
Reflections collected/unique	9950/2296
Final R indices[I>2σ(I)]	0.0355/0.0983
Goodness-of-fit on F^2	1.053
R indices (all data)	0.0412/0.0950

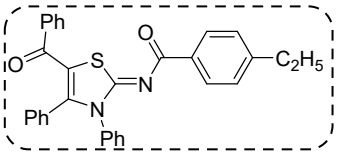
4. Characterization data of the synthesized compounds



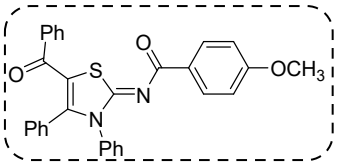
***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c1)**. Eluent: PE/EA = 4 : 1; mp: 254-255°C, pale yellow solid, yield: 87%; ^{1}H NMR (400 MHz, $CDCl_3$,) δ 8.07 (d, J = 8.2 Hz, 2H), 7.64 (d, J = 8.0 Hz, 2H), 7.45 (t, J = 7.4 Hz, 1H), 7.39-7.32 (m, 6H), 7.23-7.18 (m, 4H), 7.12-7.09 (m, 1H), 7.06-7.01 (m, 4H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$,) δ 188.6, 175.3, 168.7, 144.3, 137.6, 137.1, 136.2, 132.7, 132.0, 130.6, 129.5, 129.5, 129.2, 129.2, 129.0, 129.0, 128.7, 128.2, 128.1, 128.1, 121.3 ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{21}N_2O_2S$: 461.1318; found: 461.1314.



***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methylbenzamide (c2)**. Eluent: PE/EA = 4 : 1; mp: 244-245°C, pale yellow solid, yield: 73%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.09 - 8.05 (m, 2H), 7.62 (dd, *J* = 8.2, 1.2 Hz, 2H), 7.46 - 7.32 (m, 7H), 7.22 (ddd, *J* = 7.8, 4.9, 2.9 Hz, 4H), 6.92 (d, *J* = 8.8 Hz, 2H), 6.54 (d, *J* = 8.8 Hz, 2H), 3.66 (s, 3H) ppm. ^{13}C NMR (101 MHz, DMSO-d_6) δ 189.4, 174.4, 168.6, 145.5, 143.5, 142.8, 137.8, 137.5, 133.8, 132.7, 131.4, 129.7, 129.5, 129.5, 129.4, 129.3, 129.2, 128.4, 127.9, 121.1, 21.69 ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1430.

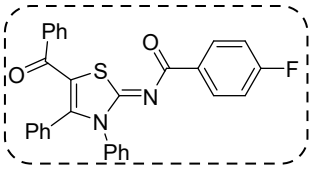


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-ethylbenzamide (c3)**. Eluent: PE/EA = 8 : 1; mp: 250-251°C, pale yellow solid, yield: 81%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.99 (d, *J* = 7.9 Hz, 2H), 7.63 (d, *J* = 7.6 Hz, 2H), 7.37 (d, *J* = 6.4 Hz, 4H), 7.20 (dd, *J* = 18.1, 7.6 Hz, 6H), 7.12 - 7.06 (m, 1H), 7.03 (d, *J* = 6.4 Hz, 4H), 2.65 (q, *J* = 7.6 Hz, 2H), 1.21 (t, *J* = 7.4 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 175.3, 168.5, 168.4, 148.8, 144.2, 137.6, 137.0, 133.9, 133.7, 133.7, 132.5, 130.6, 129.8, 129.2, 129.2, 128.9, 128.1, 127.8, 127.7, 121.3, 29.1, 15.4 ppm HRMS (ESI): m/z [M + H]+ calcd for $C_{31}H_{24}N_2O_2S$: 489.1631; found: 489.1625.

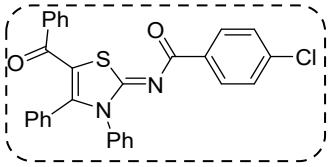


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methoxybenzamide (c4)**. Eluent: PE/EA = 4 : 1, mp: 239-240°C, pale yellow solid, yield: 82%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.96 (d, *J* = 8.9 Hz, 2H), 7.55 (d, *J* = 7.5 Hz, 2H), 7.29 (d, *J* = 6.9 Hz, 3H), 7.20 - 7.10 (m, 4H), 7.02 (t, *J* = 7.6 Hz, 2H), 6.99 - 6.91 (m, 4H), 6.83 - 6.73 (m, 2H), 3.80 (d, J = 9.4 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$, 25 °C) δ 188.8, 174.8, 168.3, 162.8, 144.2, 137.7, 137.1, 132.4, 131.6, 130.6, 129.4, 129.3, 129.2, 129.1,

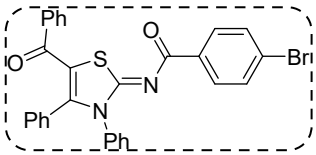
128.9, 128.9, 128.8, 128.1, 128.0, 121.2, 113.4, 55.4.ppm. HRMS (ESI): m/z [M + H]$^+$calcd for $C_{30}H_{22}N_2O_3S$: 491.1424; found: 491.1416.



***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-fluorobenzamide (c5)**. Eluent: PE/EA = 3 : 1, mp: 195-196°C, yellow solid, yield: 56%; ^{1}H NMR (400 MHz, $CDCl_3$, 25℃) δ 8.06 (dd, *J* = 8.8, 5.8 Hz, 2H), 7.64 - 7.59 (m, 2H), 7.55 - 7.44 (m, 1H), 7.41 - 7.32 (m, 3H), 7.25 - 7.18 (m, 3H), 7.15 - 7.07 (m, 2H), 7.06 - 6.95 (m, 6H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$, 25 ℃) δ 188.8, 174.7, 168.8, 144.2, 137.5, 136.9, 132.8, 132.6, 132.0 (d, *J* = 9.1 Hz), 131.9, 131.0 (d, *J* = 18.2 Hz), 130.9, 130.6, 129.6, 129.2, 128.7, 128.5, 128.1 (d, *J* = 5.1 Hz), 121.5, 115.2 (d, *J* = 20.2 Hz), ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{29}H_{19}FN_2O_2S$: 479.1224; found: 479.1197.

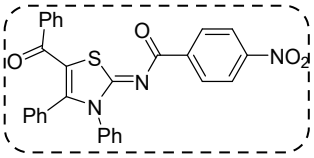


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-chlorobenzamide (c6)**. Eluent: PE/EA = 3 : 1, mp: 233-234°C, pale yellow solid, yield: 85%; ^{1}H NMR (400 MHz, $CDCl_3$, 25 ℃) δ 7.98 (d, *J* = 8.5 Hz, 2H), 7.62 (d, *J* = 7.8 Hz, 2H), 7.40 - 7.33 (m, 4H), 7.31 (d, *J* = 8.3 Hz, 2H), 7.24 - 7.17 (m, 4H), 7.11 (t, *J* = 7.1 Hz, 1H), 7.06 - 6.98 (m, 4H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$, 25 ℃) δ 188.6, 175.3, 168.6, 143.7, 137.4, 136.0, 135.4, 134.86, 132.5, 132.1, 130.6, 130.0, 129.7, 129.5, 129.2, 129.1, 128.8, 128.3, 128.2, 128.2, 121.6.ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{29}H_{19}ClN_2O_2S$: 495.0929; found: 495.0926.

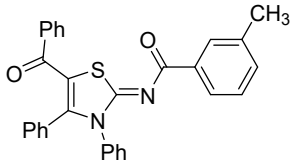


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-bromobenzamide (c7)**. Eluent: PE/EA = 3 : 1, mp: 226-227°C, yellow solid, yield: 80%; ^{1}H NMR (400 MHz, $CDCl_3$, 25 ℃) δ 13.70 (s, 1H), 7.53 (d, *J* = 7.7 Hz, 2H), 7.40 (t, *J* = 7.4 Hz, 1H), 7.31 (d, *J* = 7.6 Hz, 1H), 7.23 - 7.15 (m, 4H), 7.12 - 7.04

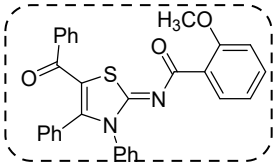
(m, 5H), 7.02 - 6.93 (m, 4H), 6.71 (d, *J* = 7.8 Hz, 1H), 6.48 (d, *J* = 7.7 Hz, 1H).ppm. HRMS (ESI): m/z $[M + Na]^+$ clack for $C_{29}H_{19}BrN_2O_2S$: 561.0243; found: 561.0219.



***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-nitrobenzamide (c8)**. Eluent: PE/EA = 4 : 1, mp: 188-189℃, yellow solid, yield: 69%; ^{1}H NMR (400 MHz, $CDCl_3$, 25 ℃) δ 8.04-8.02 (dd, *J* = 8.2, 1.4 Hz, 2H), 7.66-7.64 (dd, *J* = 8.0, 1.2 Hz, 1H), 7.64-7.61 (dd, *J* = 8.0, 1.2 Hz, 2H), 7.46-7.41 (tt, *J* = 7.2, 1.2 Hz, 1H), 7.37-7.31 (m, 4H), 7.29-7.27 (dd, *J* = 7.8, 1.8 Hz, 1H), 7.20 (t, *J* = 7.6 Hz, 2H), 7.13-7.09 (m, 3H), 7.03 (t, *J* = 7.2 Hz, 2H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$, 25 ℃) δ 188.7, 175.4, 168.1, 143.8, 137.5, 136.7, 136.1, 134.7, 133.5, 132.5, 132.1, 130.9, 130.8, 130.5, 129.8, 129.6, 129.2, 128.6, 128.3, 128.2, 128.1, 128.0, 123.3 ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{19}N_3O_4S$: 506.1169; found: 506.1136.

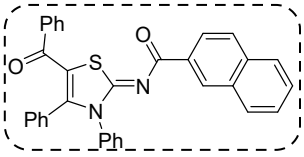


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-3-methylbenzamide (c9)**. Eluent: PE/EA = 4 : 1, mp: 224-225℃, pale yellow solid, yield: 81%; ^{13}C NMR (101 MHz, $CDCl_3$,) 180.36, 164.24, 159.88, 136.42, 128.41, 127.72, 123.69, 122.94, 122.85, 122.40, 122.32, 121.49, 120.37, 120.27, 120.22, 120.08, 119.36, 119.33, 119.05, 118.96, 112.22, 106.98, 106.76, 36.97.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1428.

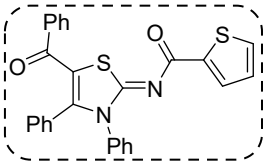


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-methoxybenzamide (c11)**. Eluent: PE/EA = 4 : 1; mp: 227-228℃, pale yellow solid, yield: 85%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.91 (d, *J* = 7.6 Hz, 1H), 7.67 (d, *J* = 7.8 Hz, 2H), 7.38 (dd, *J* = 12.3, 5.1 Hz, 5H), 7.24 (t, *J* = 7.4 Hz, 4H), 7.16 - 7.08 (m, 2H), 7.05 (d, *J* = 5.8 Hz, 3H), 6.95 (d, *J* = 8.4 Hz, 1H), 6.88 (t, *J* = 7.5 Hz, 1H), 3.90 (d, *J* = 9.4 Hz,

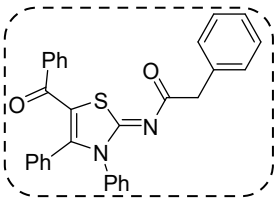
3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.6, 175.1, 168.0, 160.0, 143.9, 137.7, 137.1, 132.8, 132.5, 132.4, 132.1, 130.5, 129.3, 129.1, 128.8, 128.7, 128.7, 128.1, 127.9, 125.5, 121.1, 119.9, 112.1, 56.0.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_3S$: 491.1424; found: 491.1421.



***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-naphthamide (c13)**. Eluent: PE/EA = 4 : 1, mp: 193-194 ℃, yellow solid, yield: 76%; ^{1}H NMR (400 MHz, $CDCl_3$,) δ 8.14 (dd, *J* = 8.2, 1.5 Hz, 2H), 8.10 - 8.04 (m, 1H), 8.01 - 7.97 (m, 1H), 7.92 - 7.84 (m, 1H), 7.75 (dd, *J* = 8.6, 2.1 Hz, 1H), 7.74 - 7.69 (m, 1H), 7.62 (d, *J* = 7.3 Hz, 1H), 7.48 (t, *J* = 7.7 Hz, 5H), 7.42 – 7.32 (m, 3H), 7.24 (s, 3H), 7.08 - 7.01 (m, 1H), 6.98 -6.91 (m, 2H).ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{22}H_{17}N_2OS$: 357.1056; found: 357.1048.

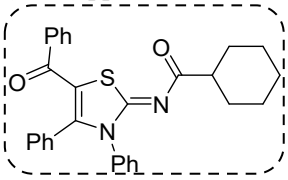


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)thiophene-2-carboxamide (c14)**. Eluent: PE/EA = 4 : 1, mp: 184-185℃, pale yellow solid, yield: 73%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.71 (d, *J* = 3.7 Hz, 1H), 7.61 (d, *J* = 7.7 Hz, 2H), 7.42 (d, *J* = 4.9 Hz, 1H), 7.36 (d, *J* = 6.5 Hz, 4H), 7.23 - 7.17 (m, 4H), 7.10 (d, *J* = 6.8 Hz, 1H), 7.02 (d, *J* = 7.5 Hz, 5H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 179.9, 178.7, 161.6, 159.3, 135.5, 133.6, 128.8, 128.0, 123.8, 123.1, 121.9, 120.8, 120.4, 120.3, 120.2, 120.2, 120.0, 119.4, 119.4, 119.2, 112.7.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{27}H_{18}N_2O_2S_2$: 467.0882; found: 467.0854.

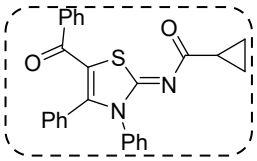


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-phenylacetamide (c15)**. Eluent: PE/EA = 10 : 1, mp: 222-223°C, pale yellow solid, yield: 44%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.41 (dd, *J* = 8.3, 1.5 Hz, 2H), 7.59 (dd, *J* = 8.3, 1.3 Hz, 2H), 7.52 (dd, *J* = 14.3, 7.8, Hz, 3H), 7.42 – 7.35 (m, 2H), 7.33

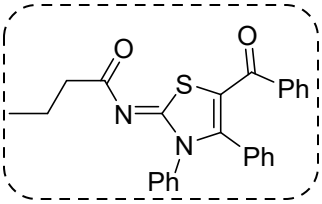
- 7.27 (m, 3H), 7.26 - 7.20 (m, 4H), 7.05 (dd, *J* = 8.2, 1.3 Hz, 2H), 6.93 (dd, *J* = 7.2, 2.2 Hz, 2H), 3.49 (s, 2H).ppm. HRMS (ESI): m/z $[M + Na]^+$ calcd for $C_{21}H_{16}NNaO$: 320.1046; found: 320.1041.



***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclohexanecarboxamide (c16)**. Eluent: PE/EA = 7 : 1; mp: 151-152°C, yellow solid, yield: 40%; ^{1}H NMR (400 MHz $CDCl_3$,) δ 7.59 (d, *J* = 7.7 Hz, 2H), 7.37 - 7.28 (m, 4H), 7.19 (t, *J* = 7.7 Hz, 2H), 7.14 - 7.09 (m, 2H), 7.07 (d, *J* = 7.3 Hz, 1H), 7.03 - 6.93 (m, 4H), 2.37 (t, *J* = 11.1 Hz, 1H), 1.86 (d, *J* = 12.8 Hz, 2H), 1.69 (d, *J* = 11.7 Hz, 2H), 1.59 (d, *J* = 8.7 Hz, 1H), 1.38 (q, *J* = 10.6, 9.9 Hz, 2H), 1.21 (tt, *J* = 17.8, 9.8 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 187.3, 168.0, 144.0, 137.7, 137.0, 132.5, 132.4, 130.6, 130.6, 129.3, 129.1, 128.8, 128.6, 128.2, 128.0, 120.9, 47.59, 29.5, 26.1, 25.9 ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{26}N_2O_2S$: 467.1788; found: 467.1779.

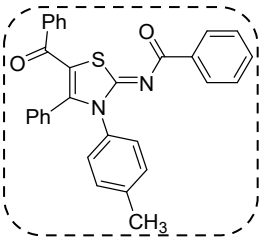


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclopropanecarboxamide (c17)**. Eluent: PE/EA = 7 : 1; mp: 148-149°C, pale yellow solid, yield: 39%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (d, *J* = 7.8 Hz, 1H), 7.64 (d, *J* = 7.6 Hz, 2H), 7.44 (t, *J* = 7.2 Hz, 1H), 7.37 (p, *J* = 7.4, 6.6 Hz, 4H), 7.22 (t, *J* = 6.7 Hz, 3H), 7.14 - 7.07 (m, 1H), 7.03 (d, *J* = 6.8 Hz, 3H), 1.27 (d, *J* = 8.7 Hz, 3H), 0.93 - 0.76 (m, 2H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 175.3, 144.2, 137.6, 137.0, 136.2, 132.6, 132.2, 132.0, 130.6, 129.6, 129.5, 129.2, 129.0, 128.7, 128.2, 128.1, 128.1, 29.8, 29.3.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{26}H_{20}N_2O_2S$: 425.1318; found: 425.1305.

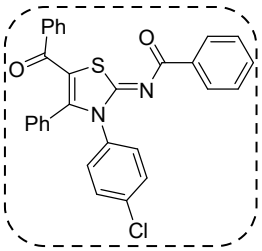


***N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)butyramide (c18)**. Eluent: PE/EA = 6: 1; mp: 136-137°C, pale yellow solid, yield: 42%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (d, *J* = 7.8 Hz, 1H), 7.64

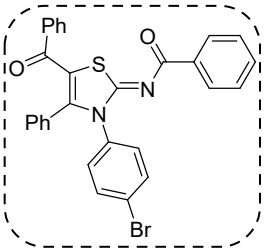
(m, 3H), 7.42 (t, *J* = 7.2 Hz, 1H), 7.36 (t, *J* = 7.4 Hz, 4H), 7.25 (t, *J* = 6.7 Hz, 3H), 7.14 - 7.07 (m, 3H), 2.58 (t, *J* = 8.0 Hz, 2H), 1.90-1.60 (m , 2H), 0.98 (t, *J* = 8.0 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 185.3, 178.2, 160.6, 147.8, 137.2, 137.0, 132.2, 128.9, 128.6, 128.6, 128.5, 128.2, 127.0, 127.8, 127.7, 122.7, 122.1, 39.1, 18.8, 13.3.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{26}H_{20}N_2O_2S$: 427.1464; found: 427.1475.



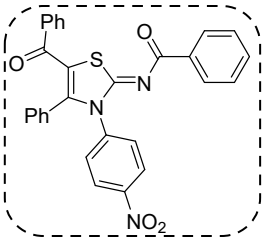
***N*-(5-benzoyl-4-phenyl-3-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c19)**. Eulent: PE/EA = 4 : 1, mp: 220-221℃, pale yellow solid, yield: 78%; ^{1}H NMR (400 MHz, $CDCl_3$,) δ 8.12 - 8.07 (m, 2H), 7.65 - 7.60 (m, 2H), 7.44 (t, *J* = 7.3 Hz, 1H), 7.35 (t, *J* = 7.7 Hz, 3H), 7.24 - 7.14 (m, 4H), 7.12 - 7.03 (m, 3H), 7.05 - 6.98 (m, 4H), 2.37 (s, 3H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 175.3, 168.8, 166.0, 164.0, 144.4, 138.9, 137.6, 136.33, 134.4, 132.5, 131.9, 130.7, 129.6, 129.5, 129.3, 129.2, 128.4, 128.1, 128.1, 121.3, 21.4.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1472.



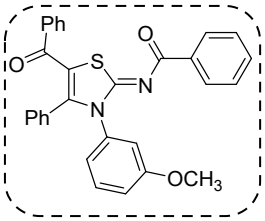
***N*-(5-benzoyl-3-(4-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c20)**. Eulent: PE/EA = 4 : 1, mp: 276-277°C, pale yellow solid, yield: 76%; ^{1}H NMR (400 MHz, $CDCl_3$) *δ* 8.09-8.07 (dd, *J* = 7.2, 1.4 Hz, 2H), 7.63-7.61 (dd, *J* = 8.4, 1.0 Hz, 2H), 7.47 (t, *J* = 7.4 Hz, 1H), 7.40-7.34 (m, 5H), 7.22 (t, *J* = 7.8 Hz, 2H), 7.19-7.13 (m, 3H), 7.08 (t, *J* = 7.6 Hz, 2H), 7.02-7.00 (m, 2H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.6, 175.3, 168.7, 143.7, 137.4, 136.0, 135.5, 134.9, 132.6, 132.2, 130.6, 130.1, 129.7, 129.6, 129.2, 129.2, 128.9, 128.3, 128.2, 128.2, 121.6 ppm. HRMS(ESI) m/z $[M + H]^+$ calcd for $C_{29}H_{20}ClN_2O_2S$, 495.0934, found 495.0931.



***N*-(5-benzoyl-3-(4-bromophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c21)**. Eluent: PE/EA = 4 : 1, mp: 265-266°C, yellow solid, yield: 80%; ^{13}C NMR (101 MHz, $CDCl_3$, 25 ℃) δ 188.6, 175.3, 168.6, 143.6, 137.4, 136.0, 132.6, 132.2, 130.6, 130.4, 130.3, 129.8, 129.6, 129.2, 128.9, 128.6, 128. 3, 128.2, 128.2, 123.0, 121.6.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{19}BrN_2O_2S$: 529.0423; found: 529.0413.

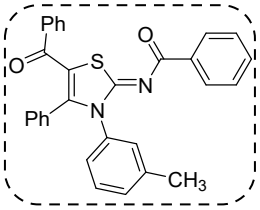


***N*-(5-benzoyl-3-(4-nitrophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c22)**. Eluent: PE/EA = 5 : 1, mp: 187-188°C, pale yellow solid, yield: 66%; ^{1}H NMR (400 MHz, $CDCl_3$, 25 ℃) δ 8.26 (d, *J* = 9.1 Hz, 2H), 8.04 (dd, *J* = 7.8, 1.4 Hz, 2H), 7.66 - 7.60 (m, 2H), 7.45 (d, *J* = 8.7 Hz, 3H), 7.41 -7.35 (m, 3H), 7.23 (t, *J* = 7.9 Hz, 2H), 7.16 (d, *J* = 7.4 Hz, 1H), 7.09 (t, *J* = 7.6 Hz, 2H), 7.04 - 7.00 (m, 2H).ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{19}N_3O_4S$: 506.1169; found: 506.1166.

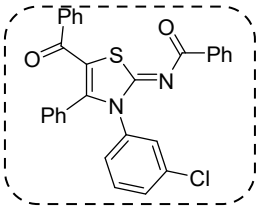


***N*-(5-benzoyl-3-(3-methoxyphenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c23)**. Eluent: PE/EA = 5 : 1; pale yellow solid, mp: 205-206°C, yield: 84%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.10 (dd, *J* = 8.2, 1.3 Hz, 2H), 7.63 (dd, *J* = 8.2, 1.3 Hz, 2H), 7.45 (t, *J* = 6.7 Hz, 1H), 7.36 (t, *J* = 7.5 Hz, 3H), 7.29 (d, *J* = 8.2 Hz, 2H), 7.22 (t, *J* = 7.7 Hz, 2H), 7.12 (dt, *J* = 5.6, 3.1 Hz, 1H), 7.07 - 7.03 (m, 3H), 6.90 (dd, *J* = 8.5, 2.4 Hz, 1H), 6.84 (dd, *J* = 8.0, 1.9 Hz, 1H), 6.74 (t, *J* = 2.2 Hz, 1H), 3.71 (s, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.7, 175.3, 168.6, 159.9, 144.1, 137.9, 137.6, 136.2, 132.6, 132.0,

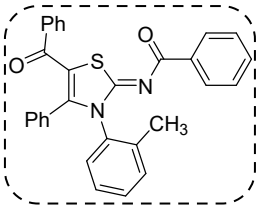
130.5, 129.6, 129.6, 129.6, 129.3, 129.2, 128.2, 128.1, 121.3, 121.1, 115.2, 114.2, 55.6. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{23}N_2O_3S$: 491.1424; found: 491.1418.



***N*-(5-benzoyl-4-phenyl-3-(m-tolyl)thiazol-2(3H)-ylidene)benzamide (c24)**. Eluent: PE/EA = 6 : 1; mp: 224-225°C, pale yellow solid, yield: 79%; ^{1}H NMR (400 MHz, $CDCl_3$,) δ 7.63 (dd, *J* = 8.3, 1.2 Hz, 2H), 7.44 (t, *J* = 7.4 Hz, 1H), 7.38 - 7.32 (m, 3H), 7.26 - 7.19 (m, 3H), 7.17 - 7.08 (m, 2H), 7.07 - 6.99 (m, 6H), 2.30 (s, 3H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.7, 175.3, 168.7, 144.3, 138.9, 137.6, 136.9, 136.3, 132.5, 131.9, 130.6, 129.7, 129.6, 129.5, 129.3, 129.2, 128.7, 128.6, 128.2, 128.0, 128.0, 125.8, 121.3, 21.3.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1480.

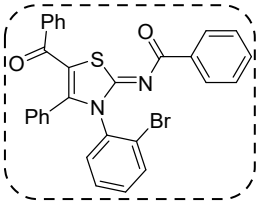


***N*-(5-benzoyl-3-(3-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c25)**. Eluent: PE/EA = 4 : 1, mp: 241-242°C, pale yellow solid, yield: 81%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.14 - 8.07 (m, 3H), 7.91 (ddd, *J* = 22.4, 8.5, 1.5 Hz, 1H), 7.62 (td, *J* = 7.8, 7.3, 1.5 Hz, 3H), 7.54 - 7.43 (m, 4H), 7.40 - 7.33 (m, 2H), 7.34 - 7.29 (m, 1H), 7.22 (t, *J* = 7.8 Hz, 1H), 7.18 - 7.13 (m, 1H), 7.11 - 7.01 (m, 3H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.6, 175.3, 168.6, 143.6, 137.9, 137.4, 136.0, 134.5, 133.7, 132.6, 132.2, 130.6, 130.3, 129.9, 129.8, 129.6, 129.3, 129.2, 128.3, 128.2, 128.2, 127.1, 121.6.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{19}ClN_2O_2S$: 495.0929; found: 495.0928.

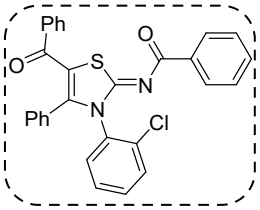


***N*-(5-benzoyl-4-phenyl-3-(o-tolyl)thiazol-2(3H)-ylidene)benzamide (c26)**. Eluent: PE/EA = 5 :

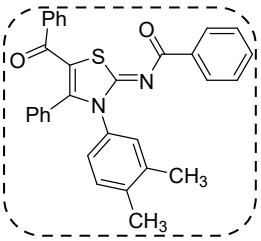
1; mp: 216-217°C, pale yellow solid, yield: 72%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.04-8.02 (dd, J = 8.2, 1.4 Hz, 2H), 7.64-7.62 (dd, J = 7.8, 1.4 Hz, 2H), 7.45-7.41 (tt, J = 7.2, 1.6 Hz, 1H), 7.37-7.30 (m, 3H), 7.29-7.21 (m, 2H), 7.21-7.16 (m, 3H), 7.11-7.06 (m, 2H), 7.02-7.00 (m, 4H), 2.16 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 175.3, 168.0, 144.4, 137.6, 136.4, 136.2, 136.0, 132.5, 132.0, 131.0, 130.3, 130.1, 129.6, 129.6, 129.5, 129.2, 128.9, 128.5, 128.2, 128.0, 126.7, 121.5, 18.1; HRMS(ESI) m/z [M + H]$^+$ calcd for $C_{30}H_{23}N_2O_2S$, 475.1475, found 475.1469.



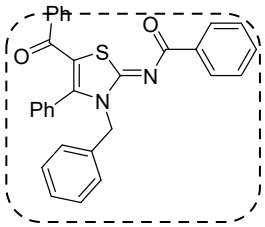
***N*-(5-benzoyl-3-(2-bromophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c27)**. Eluent: PE/EA = 4 : 1; mp: 235-236°C, pale yellow solid, yield: 65%; ^{1}H NMR (400 MHz, $CDCl_3$,) δ 8.04-8.02 (dd, J = 8.2, 1.4 Hz, 2H), 7.66-7.64 (dd, J = 8.0, 1.2 Hz, 1H), 7.65-7.62 (dd, J = 8.0, 1.2 Hz, 2H), 7.46-7.41 (tt, J = 7.2, 1.2 Hz, 1H), 7.37-7.31 (m, 4H), 7.29-7.27 (dd, J = 7.8, 1.8 Hz, 1H), 7.20 (t, J = 7.6 Hz, 2H), 7.13-7.09 (m, 3H), 7.03 (t, J = 7.2 Hz, 2H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$,) δ 188.7, 175.3, 168.1, 143.9, 137.5, 136.6, 136.1, 134.7, 133.4, 132.5, 132.1, 130.9, 130.8, 130.7, 129.8, 129.6, 129.2, 128.6, 128.4, 128.2, 128.1, 128.2, 123.3 ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{29}H_{20}BrN_2O_2S$: 539.0423; found: 539.0416.



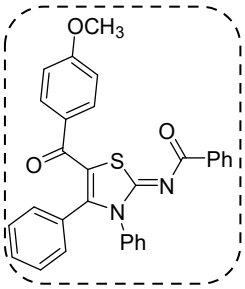
***N*-(5-benzoyl-3-(2-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c28)**. Eluent: PE/EA = 4 : 1, mp: 230-231°C, pale yellow solid, yield: 80%; ^{1}H NMR (400 MHz, $CDCl_3$, 25 °C) δ 8.10 (d, J = 7.2 Hz, 2H), 7.63 (d, J = 8.0 Hz, 2H), 7.44 (t, J = 7.4 Hz, 1H), 7.37-7.34 (m, 3H), 7.21 (t, J = 7.8 Hz, 2H), 7.13-7.10 (m, 2H), 7.06-7.01 (m, 4H), 6.96-6.94 (m, 2H), 2.26 (s, 3H), 2.19 (s, 3H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.7, 175.4, 168.2, 143.9, 137.5, 136.2, 135.1, 133.1, 132.5, 132.1, 130.8, 130.7, 130.3, 130.3, 129.8, 129.6, 129.2, 128.6, 128.2, 128.1, 128.0, 127.6, 121.5.ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{29}H_{19}ClN_2O_2S$: 495.0929; found: 495.0934.



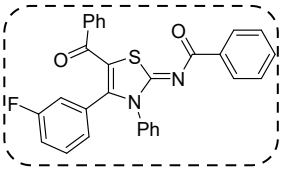
***N*-(5-benzoyl-3-(3,4-dimethylphenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c29)**. Eluent: PE/EA = 4 : 1; mp: 224-225℃, pale yellow solid, yield: 76%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 (d, J = 7.2 Hz, 2H), 7.62 (d, J = 8.0 Hz, 2H), 7.45 (t, J = 7.4 Hz, 1H), 7.37-7.33 (m, 3H), 7.20 (t, J = 7.8 Hz, 2H), 7.13-7.10 (m, 2H), 7.06-7.02 (m, 4H), 6.96-6.94 (m, 2H), 2.26 (s, 3H), 2.19 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 175.3, 168.8, 144.5, 137.7, 137.5, 137.4, 136.4, 134.5, 132.6, 131.9, 130.5, 130.0, 129.7, 129.5, 129.4, 129.3, 129.2, 128.1 128.0, 125.7, 121.3, 115.2, 19.8, 19.7; HRMS(ESI) m/z [M + H]$^+$ calcd for $C_{31}H_{25}N_2O_2S$, 489.1631, found 489.1627.



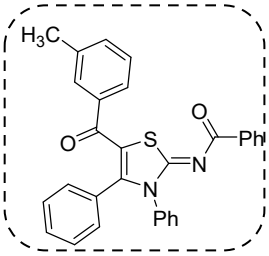
***N*-(5-benzoyl-3-benzyl-4-phenylthiazol-2(3H)-ylidene)benzamide (c31)**. Eluent: PE/EA = 4 : 1, mp: 216-217℃, pale yellow solid, yield: 58%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.41 (dd, J = 8.3, 1.5 Hz, 2H), 7.59 (dd, J = 8.3, 1.3 Hz, 2H), 7.52 (ddd, J = 14.3, 7.8, 6.2 Hz, 3H), 7.42 - 7.35 (m, 2H), 7.33 - 7.27 (m, 3H), 7.26 - 7.20 (m, 4H), 7.05 (dd, J = 8.2, 1.3 Hz, 2H), 6.93 (dd, J = 7.2, 2.2 Hz, 2H), 3.49 (s, 2H).ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1435.



***N*-(5-(4-methoxybenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c32)**. Eluent: PE/EA = 4 : 1; mp: 226-227°C, yellow solid, yield: 87%; ^{1}H NMR (400 MHz,$CDCl_3$) δ 8.08 (d, *J* = 7.7 Hz, 2H), 7.72 (d, *J* = 8.8 Hz, 2H), 7.51 - 7.41 (m, 2H), 7.39 - 7.31 (m, 4H), 7.23 (dd, *J* = 7.6, 2.0 Hz, 2H), 7.16 - 7.11 (m, 1H), 7.10 - 7.02 (m, 4H), 6.74 (d, *J* = 8.8 Hz, 2H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.9, 175.3, 168.8, 160.3, 144.3, 137.7, 137.2, 136.3, 132.4, 132.1, 131.9, 129.6, 129.2, 129.0, 128.9, 128.9, 128.1, 121.1, 120.9, 113.6, 109.7, 55.3.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_3S$: 491.1424; found: 491.1427.

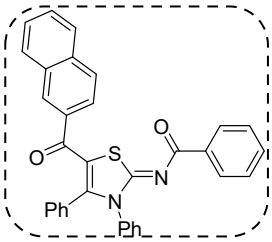


***N*-(5-benzoyl-4-(3-fluorophenyl)-3-phenylthiazol-2(3H)-ylidene)benzamide (c33)**. Eluent: PE/EA = 4 : 1; mp: 168-169°C, pale yellow solid, yield: 82%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.07 (d, *J* = 7.8 Hz, 2H), 7.67 (d, *J* = 7.8 Hz, 2H), 7.48 - 7.38 (m, 5H), 7.35 (t, *J* = 7.5 Hz, 2H), 7.30 - 7.26 (m, 2H), 7.23 (dd, *J* = 7.5, 2.1 Hz, 2H), 7.04 (td, *J* = 8.0, 5.8 Hz, 1H), 6.87 - 6.80 (m, 2H), 6.76 (d, *J* = 8.9 Hz, 1H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$, 25 °C) δ 188.4, 175.3, 168.5, 162.0 (d, *J* = 249.5 Hz), 160.7, 142.5, 137.6, 136.8, 136.0, 132.8, 132.1, 131.2 (d, *J* = 8.1 Hz), 129.8 (d, *J* = 8.1 Hz), 129.6, 129.2, 129.1 (d, *J* = 3.0 Hz), 128.6, 128.3, 128.2 (d, *J* = 15.2 Hz), 126.5 (d, *J* = 3.0 Hz), 121.7, 117.9, 116.7, ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{29}H_{19}FN_2O_2S$: 479.1224; found: 479.1240.

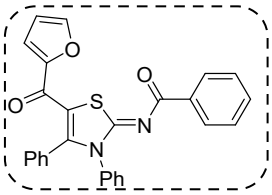


***N*-(5-(3-methylbenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c34)**. Eluent: PE/EA = 4 : 1, mp: 223-224°C, yellow solid, yield: 82%. ^{1}H NMR (400 MHz, $CDCl_3$, 25 °C) δ 8.09 - 8.05 (m, 2H), 7.50 - 7.41 (m, 2H), 7.40 - 7.32 (m, 6H), 7.22 (dd, *J* = 7.5, 2.1 Hz, 2H), 7.18 - 7.07 (m, 3H), 7.03 (d, *J* = 7.7 Hz, 4H), 2.22 (s, 3H).ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.9, 175.3, 168.7, 143.9, 137.8, 137.5, 137.1, 136.3, 133.3, 132.0, 130.6, 129.8, 129.6, 129.5, 129.3, 128.9, 128.9, 128.8, 128.1, 128.0, 126.4, 121.7, 21.2.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found:

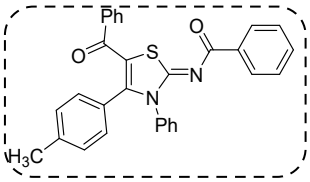
475.1484.



***N*-(5-(2-naphthoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c35)**. Eluent: PE/EA = 5 : 1, mp: 186-187℃, yellow solid, yield: 84%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 (dd, *J* = 8.2, 1.5 Hz, 2H), 8.10 - 8.04 (m, 1H), 8.00 - 7.97 (m, 1H), 7.92 -7.84 (m, 1H), 7.76 (dd, *J* = 8.6, 2.1 Hz, 1H), 7.73 - 7.69 (m, 1H), 7.61 (d, *J* = 7.3 Hz, 1H), 7.48 (t, *J* = 7.7 Hz, 5H), 7.40 - 7.32 (m, 3H), 7.21 (s, 3H), 7.09 - 7.01 (m, 1H), 6.97 - 6.91 (m, 2H).ppm ^{13}C NMR (101 MHz, $CDCl_3$) δ 191.6, 182.9, 178.9, 161.9, 152.6, 148.2, 146.1, 140.8, 135.4, 133.9, 132.0, 130.5, 130.3, 129.6, 129.5, 129.4, 128.9, 128.8, 128.6, 128.3, 128.1, 128.1, 127.8, 127.1, 126.7, 126.6, 124.7.ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{33}H_{22}N_2O_2S$: 511.1475; found: 511.1485.

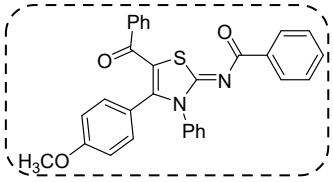


***N*-(5-(furan-2-carbonyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c36)**. Eluent: PE/EA = 8 : 1, mp: 178-179℃, pale yellow solid, yield: 80%.^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08 (d, *J* = 7.6 Hz, 2H), 7.73 (d, *J* = 3.8 Hz, 1H), 7.62 (d, *J* = 4.9 Hz, 1H), 7.46 (t, *J* = 7.3 Hz, 1H), 7.40 - 7.32 (m, 5H), 7.24 - 7.19 (m, 3H), 7.16 (d, *J* = 6.8 Hz, 4H), 7.03 - 6.99 (m, 1H).ppm. HRMS (ESI): m/z [M + H]$^+$ calcd for $C_{27}H_{18}N_2O_3S$: 451.1111; found: 451.1125.

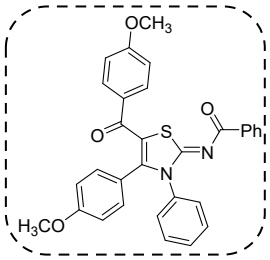


***N*-(5-benzoyl-3-phenyl-4-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c37)**. Eluent: PE/EA = 4 : 1, mp: 230-231°C, pale yellow solid, yield: 80%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.12 - 8.07 (m, 2H), 7.64 - 7.61 (m, 2H), 7.43 (d, *J* = 7.3 Hz, 1H), 7.35 (t, *J* = 7.7 Hz, 3H), 7.23 - 7.13 (m, 4H), 7.09 (d, *J* =

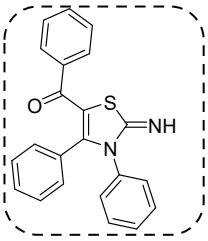
8.5 Hz, 3H), 7.07 - 6.98 (m, 4H), 2.38 (s, 3H).^{13}C NMR (101 MHz, $CDCl_3$) δ 188.8, 176.6, 175.3, 168.8, 164.0, 144.4, 138.9, 137.6, 136.3, 134.4, 132.5, 131.9, 130.7, 129.6, 129. 5, 129.2, 129.1, 128.4, 128.1, 128.1, 121.3, 21.4.ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{30}H_{22}N_2O_2S$: 475.1475; found: 475.1430.



***N*-(5-benzoyl-4-(4-methoxyphenyl)-3-phenylthiazol-2(3H)-ylidene)benzamide (c37-1)**. Eluent: PE/EA = 3 : 1; mp = 195-196°C, pale yellow solid, yield: 82%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.08-8.05 (m, 2H), 7.65- 7.61 (dd, J = 8.0, 1.2 Hz, 2H), 7.45-7.33 (m, 7H), 7.24-7.20 (m, 4H), 6.91 (d, J = 8.8 Hz, 2H), 6.54 (d, J = 8.8 Hz, 2H), 3.66 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 188.9, 175.3, 168.8, 160.3, 144.3, 137.7, 137.2, 136.3, 132.4, 132.1, 132.0, 129.6, 129.2, 129.1, 128.9, 128.8, 128.2, 121.1, 120.8, 113.7, 109.6, 55.3 ppm. HRMS(ESI) m/z $[M + H]^+$ calcd for $C_{30}H_{23}N_2O_3S$, 491.1424, found 491.1415.



***N*-(5-(4-methoxybenzoyl)-4-(4-methoxyphenyl)-3-phenylthiazol-2(3H)-ylidene)benzamide (c38)**. Eluent: PE/EA = 3 : 1; mp: 172-173°C, pale yellow solid, yield: 77%; ^{1}H NMR (400 MHz, $CDCl_3$) δ 8.09 - 8.05 (m, 1H), 7.71 (d, J = 8.7 Hz, 1H), 7.44 - 7.37 (m, 2H), 7.34 (s, 1H), 7.25 - 7.20 (m, 1H), 6.95 (d, J = 9.1 Hz, 1H), 6.74 (d, J = 9.1 Hz, 1H), 6.58 (d, J = 8.6 Hz, 1H), 3.79 (s, 2H), 3.67 (s, 2H) ppm. ^{13}C NMR (101 MHz, $CDCl_3$) δ 187.3, 175.1, 168.6, 163.3, 160.3, 143.0, 137.3, 136.4, 132.0, 131.9, 131.9, 130.2, 129.6, 129.0, 128.9, 128.8, 128.1, 121.3, 120.9, 113.7, 113.6, 55.6, 55.3 ppm. HRMS (ESI): m/z $[M + H]^+$ calcd for $C_{31}H_{24}N_2O_4S$: 521.1530; found: 521.1530.



(2-imino-3,4-diphenyl-2,3-dihydrothiazol-5-yl)(phenyl)methanone (imine). Eluent: PE/EA = 5 : 1; mp: 192-193°C, yellow solid. ^{1}H NMR (400 MHz, $CDCl_3$) δ 7.48 (d, J = 8.0 Hz, 2H), 7.31 (d, J = 8.4 Hz, 2H), 7.26-7.18 (m, 3H), 7.11-7.10 (m, 8H); ^{13}C NMR (101 MHz, $CDCl_3$) δ 189.0, 168.8, 158.1, 139.2, 138.0, 134.5, 131.7, 129.9, 129.6, 129.3, 128.9, 127.9, 127.8, 124.6, 122.1, 119.9; HRMS(ESI) m/z $[M + H]^+$ calcd for $C_{22}H_{17}N_2OS$, 357.1056, found 357.1048.

5. NMR spectra of the synthesized compounds

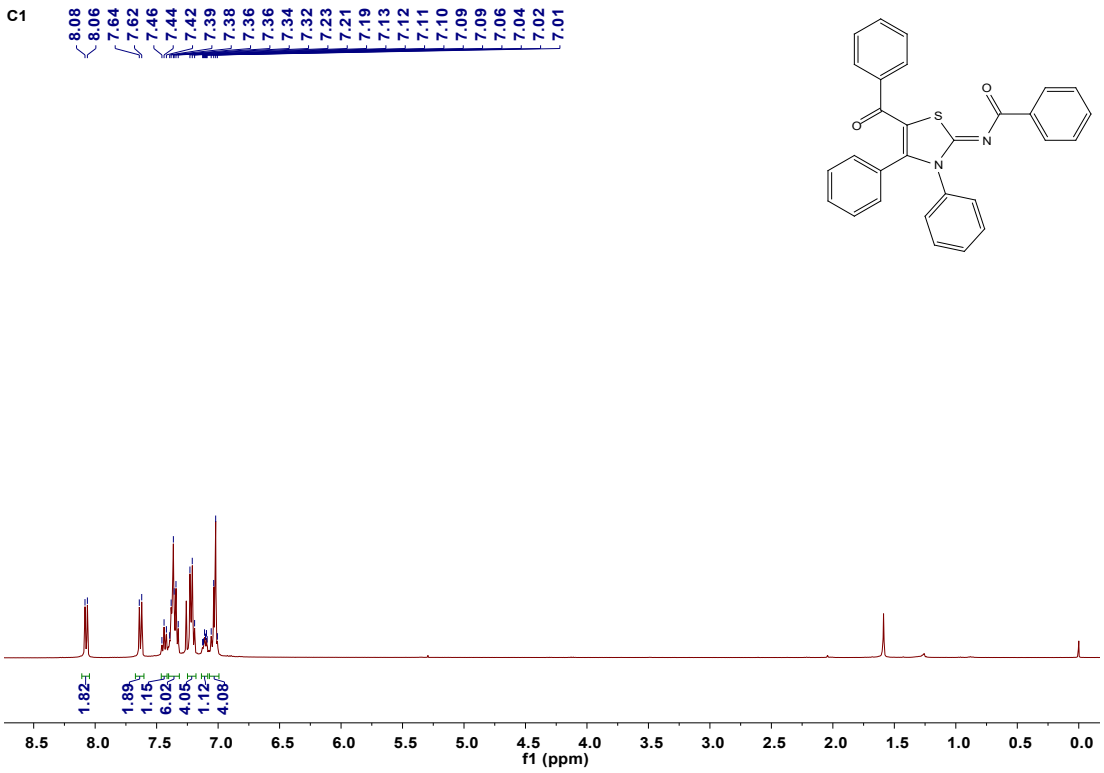


Figure S5 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c1)

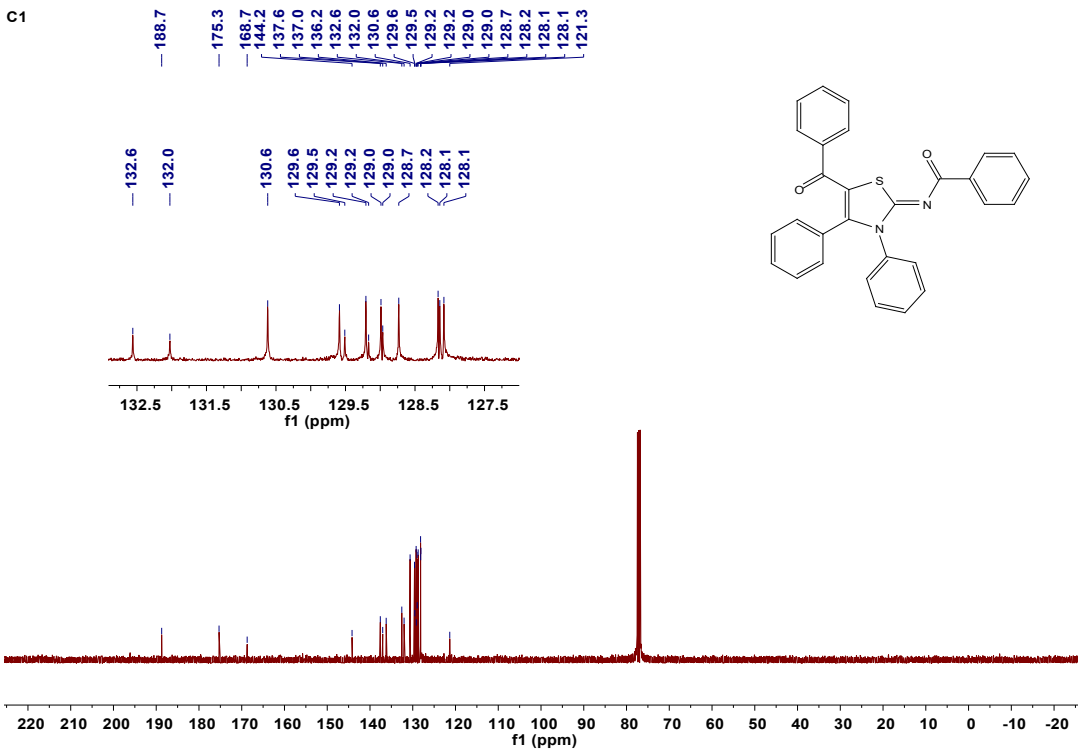


Figure S6 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c1)

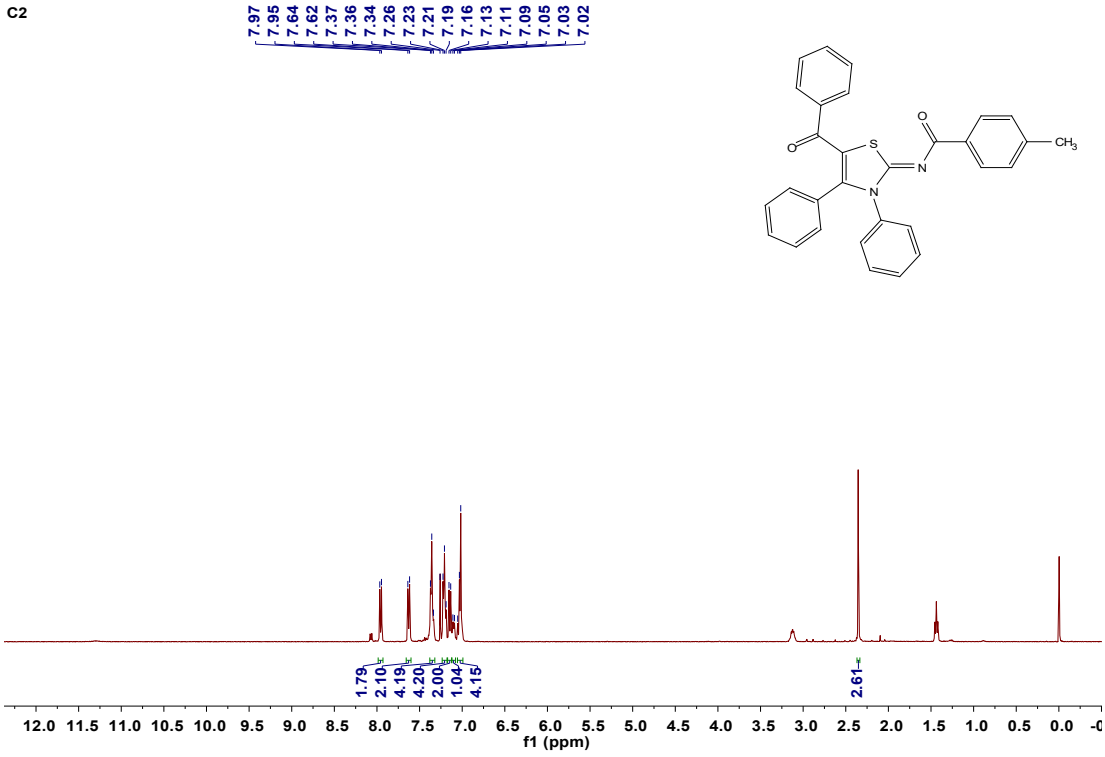


Figure S7 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methylbenzamide (c2)

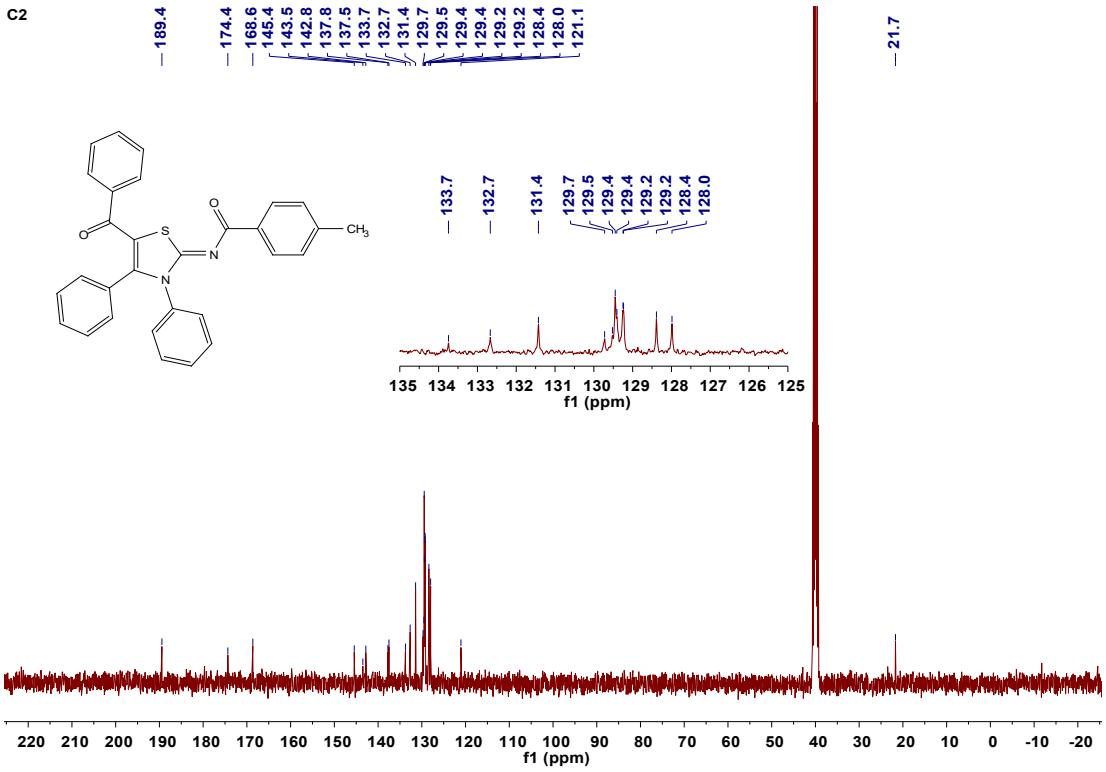


Figure S8 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methylbenzamide (c2)

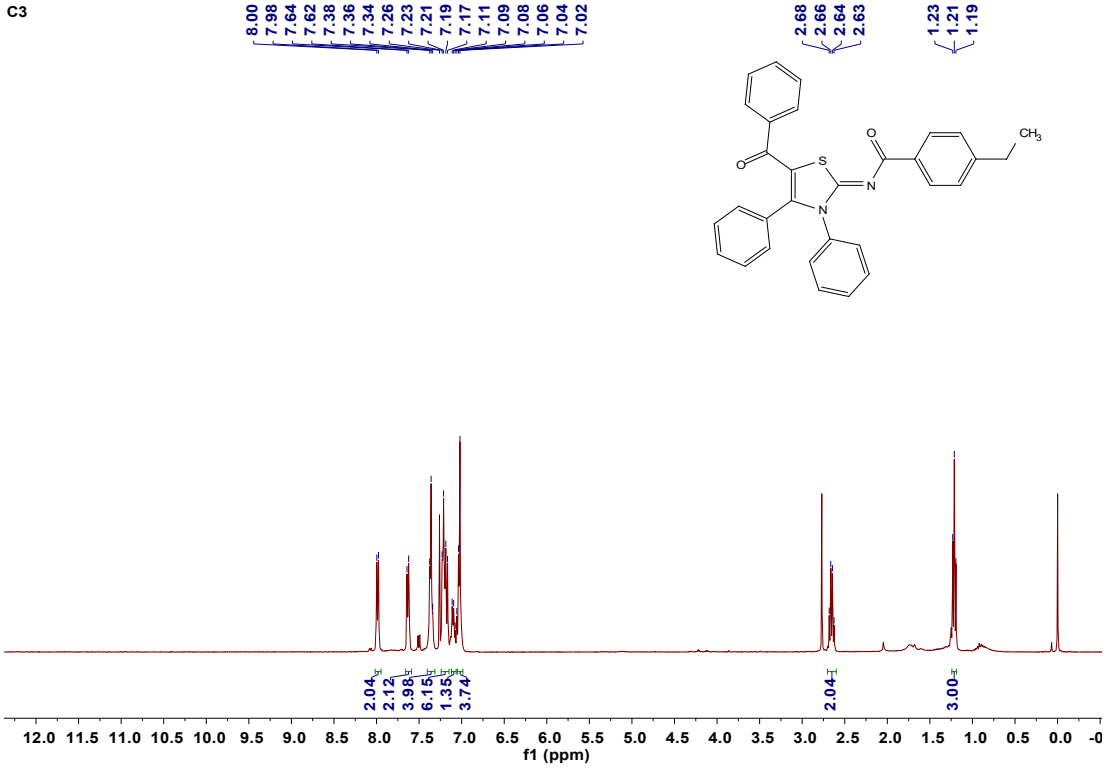


Figure S9 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-ethylbenzamide (c3)

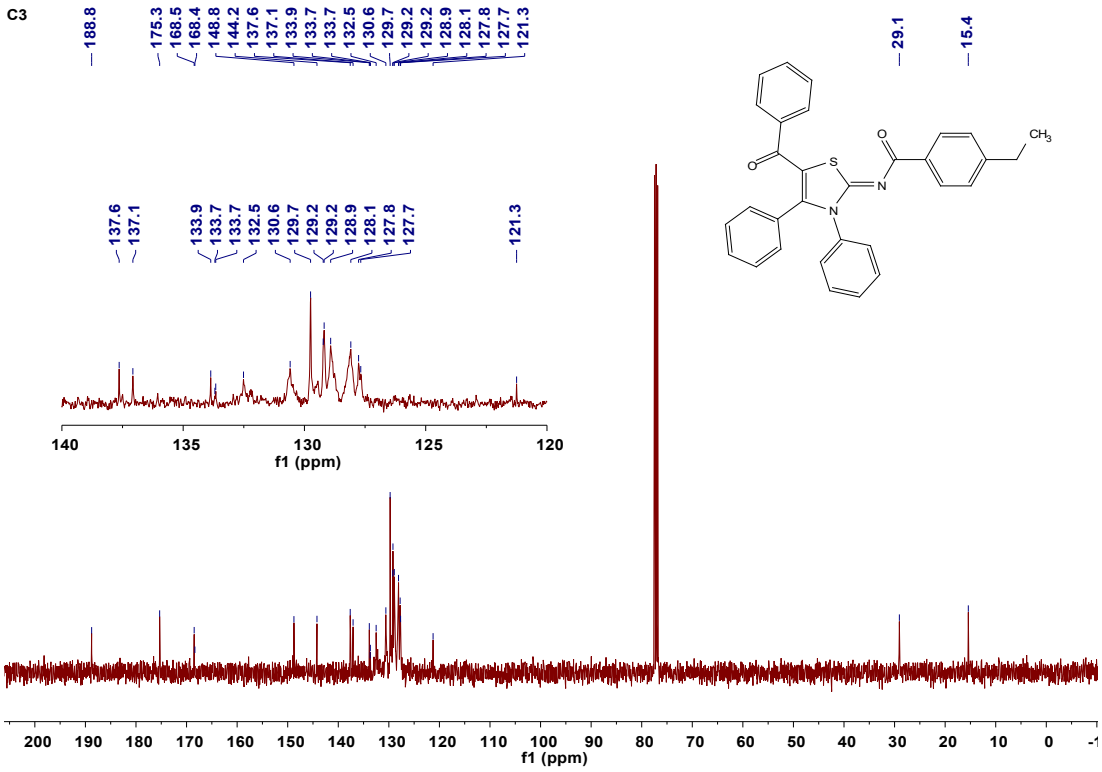


Figure S10 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-ethylbenzamide (c3)

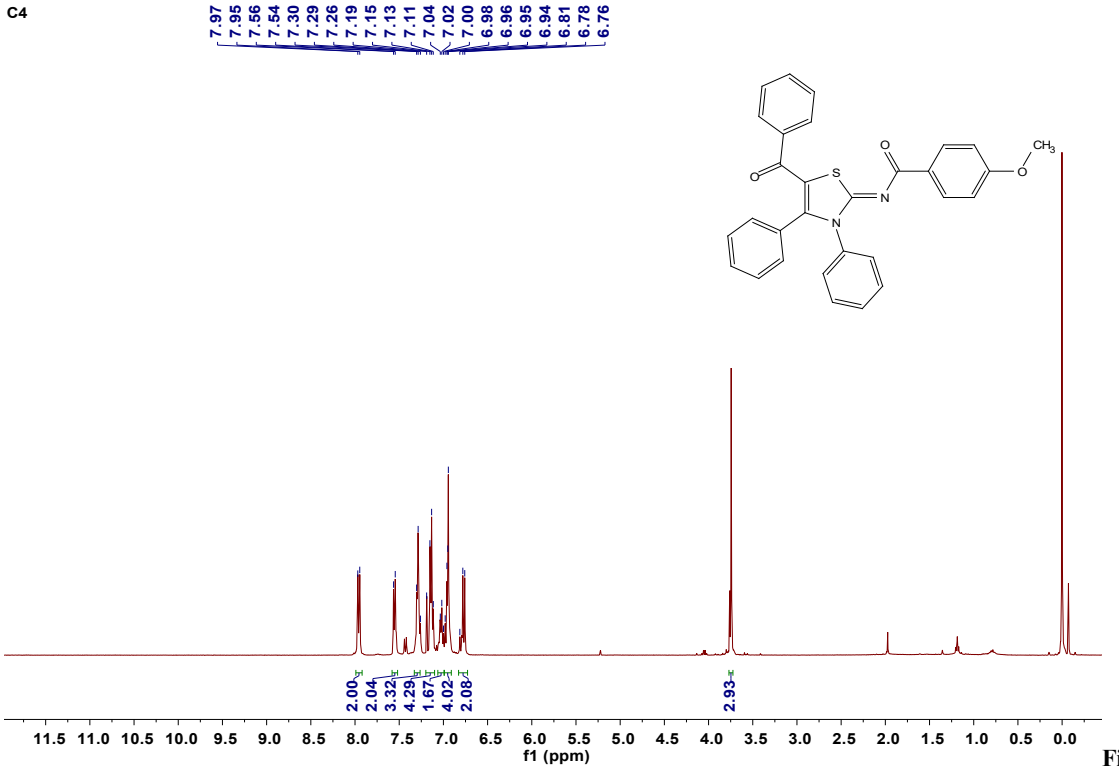


Figure S11 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methoxybenzamide (c4)

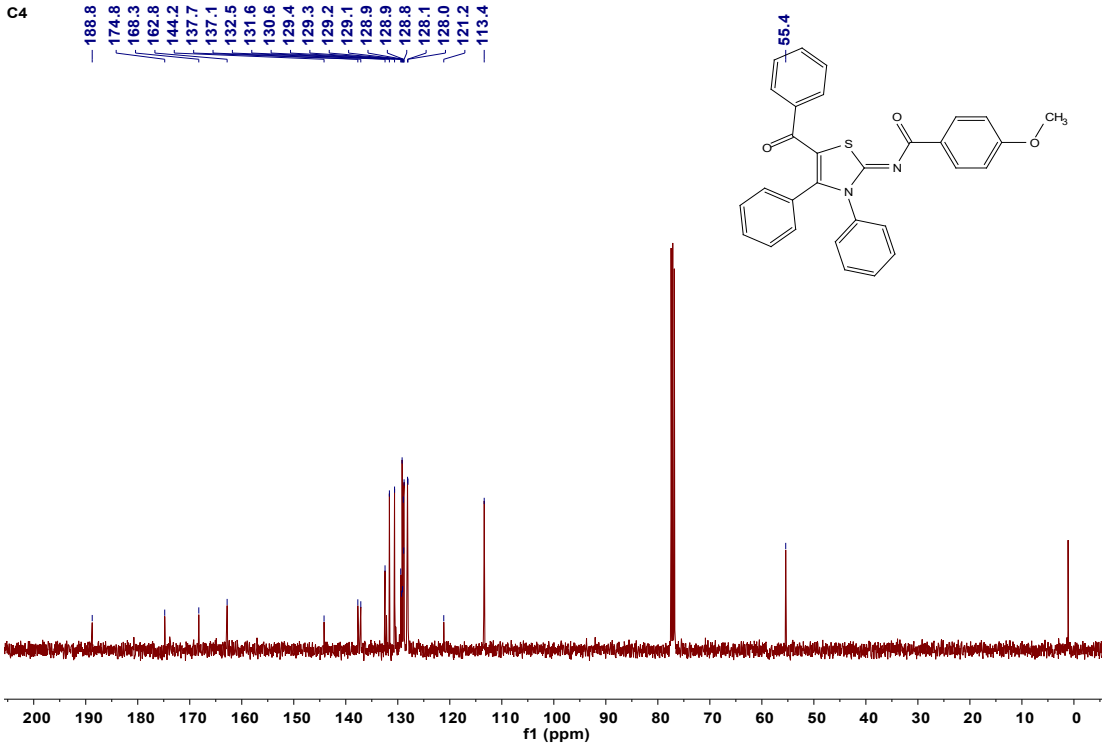


Figure S12 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-methoxybenzamide (c4)

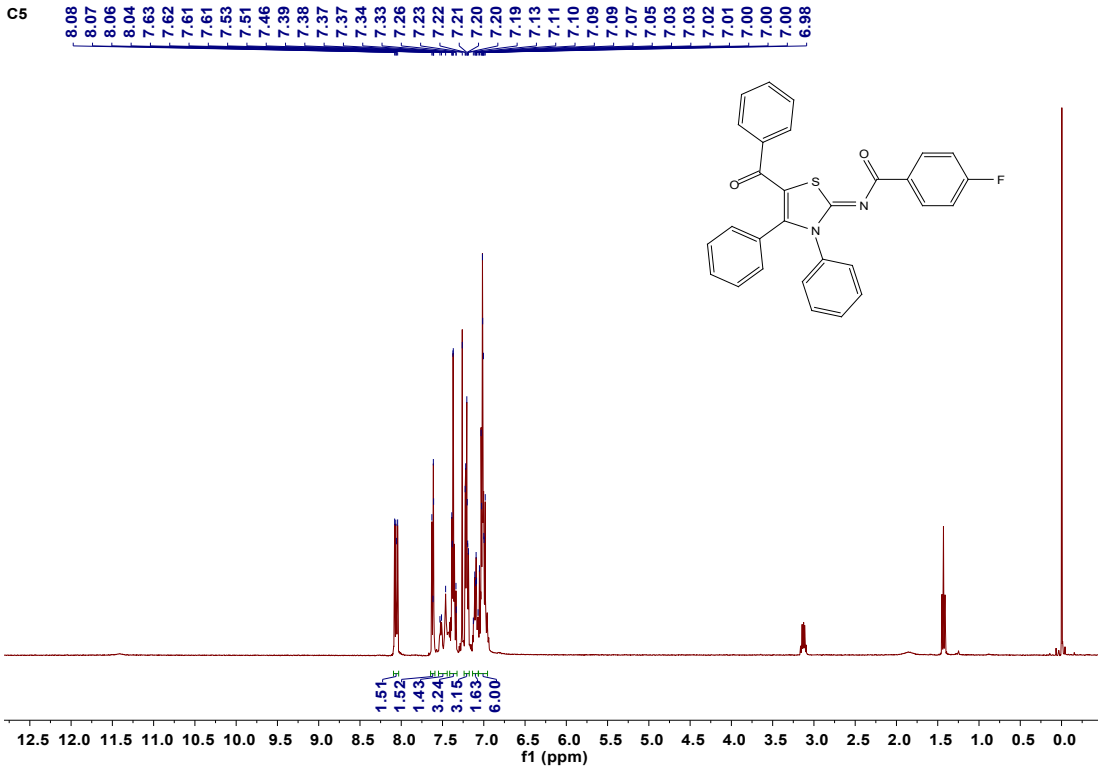


Figure S13 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-fluorobenzamide (c5)

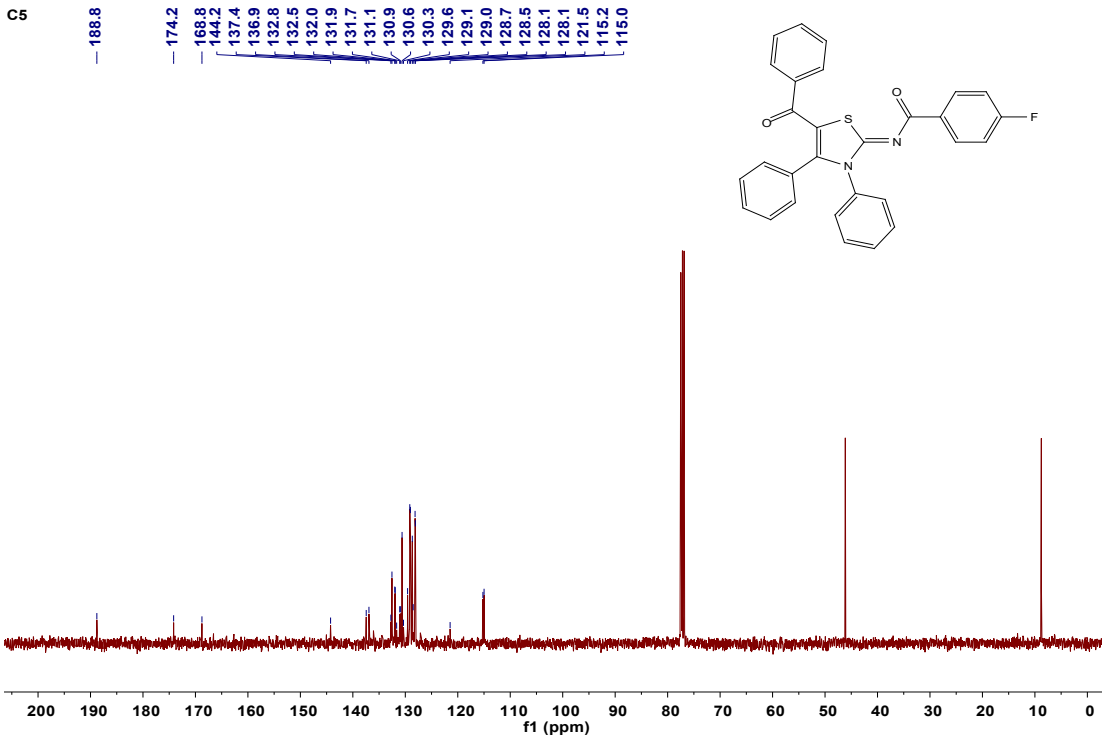


Figure S14 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-fluorobenzamide (c5)

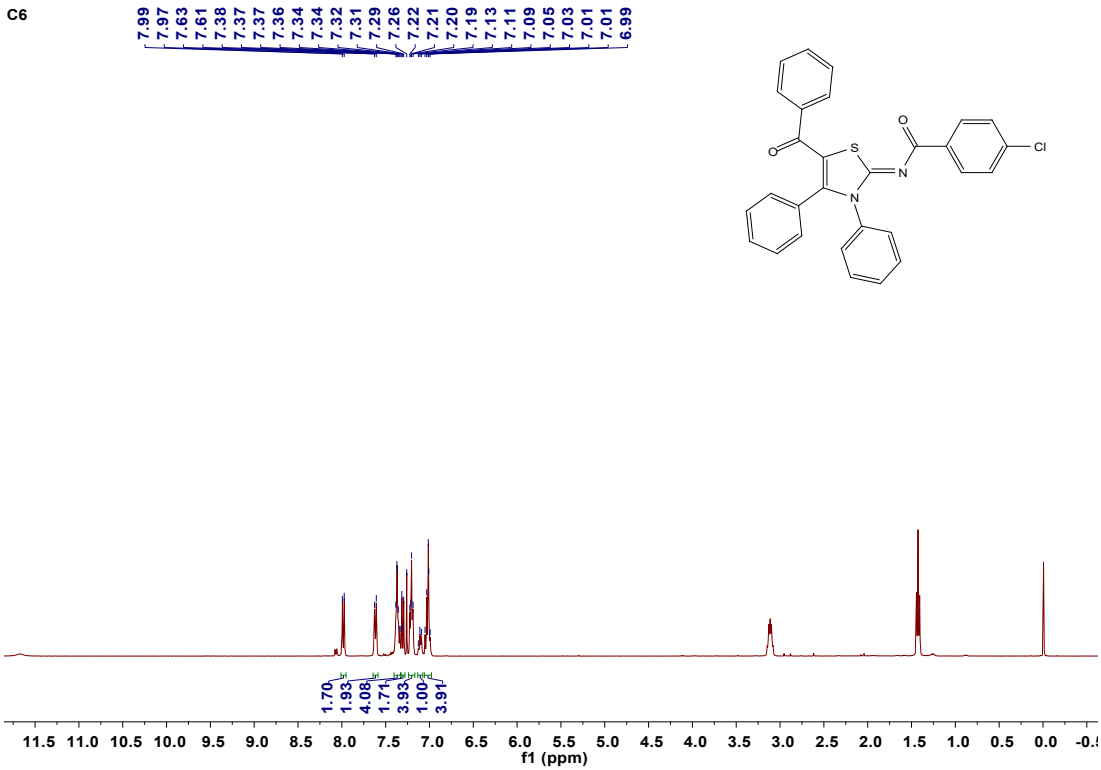


Figure S15 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-chlorobenzamide (c6)

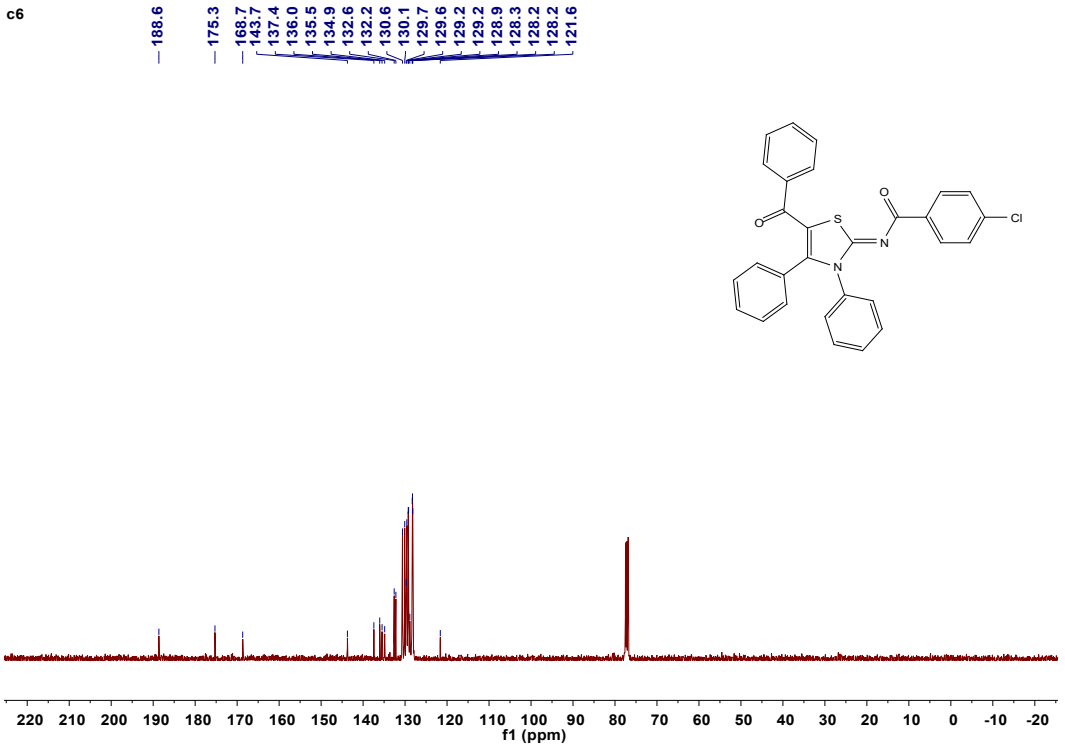


Figure S16 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-chlorobenzamide (c6)

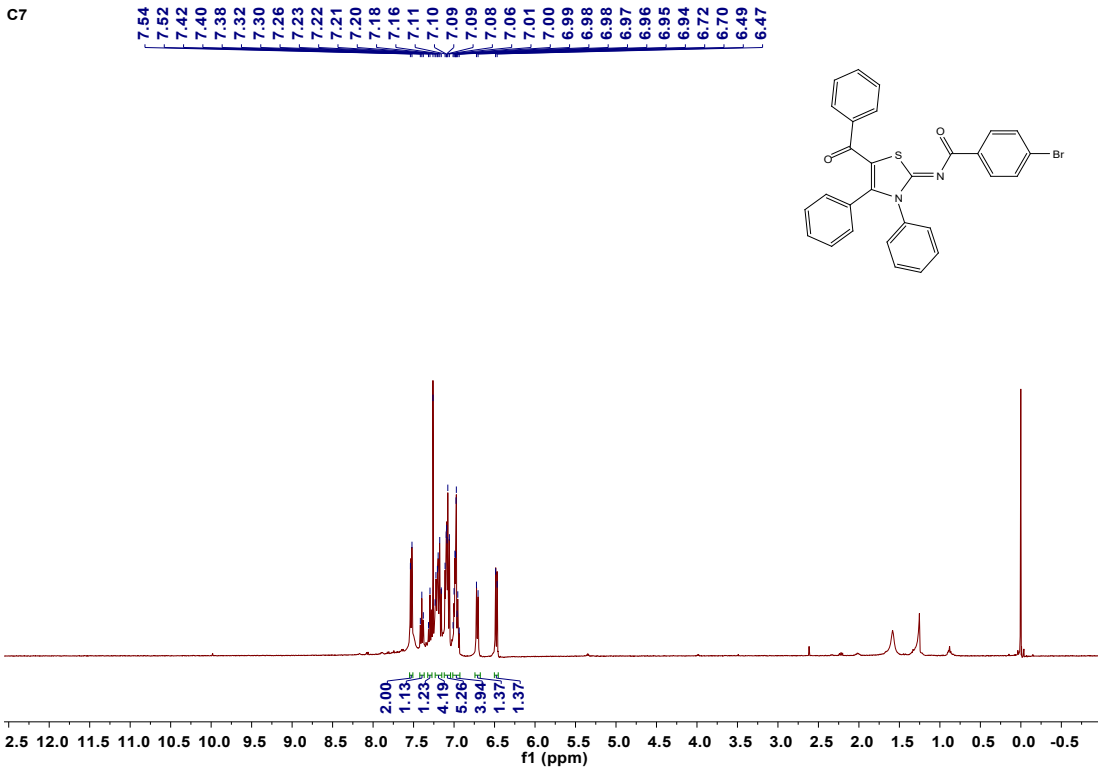


Figure S17 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-bromobenzamide (c7)

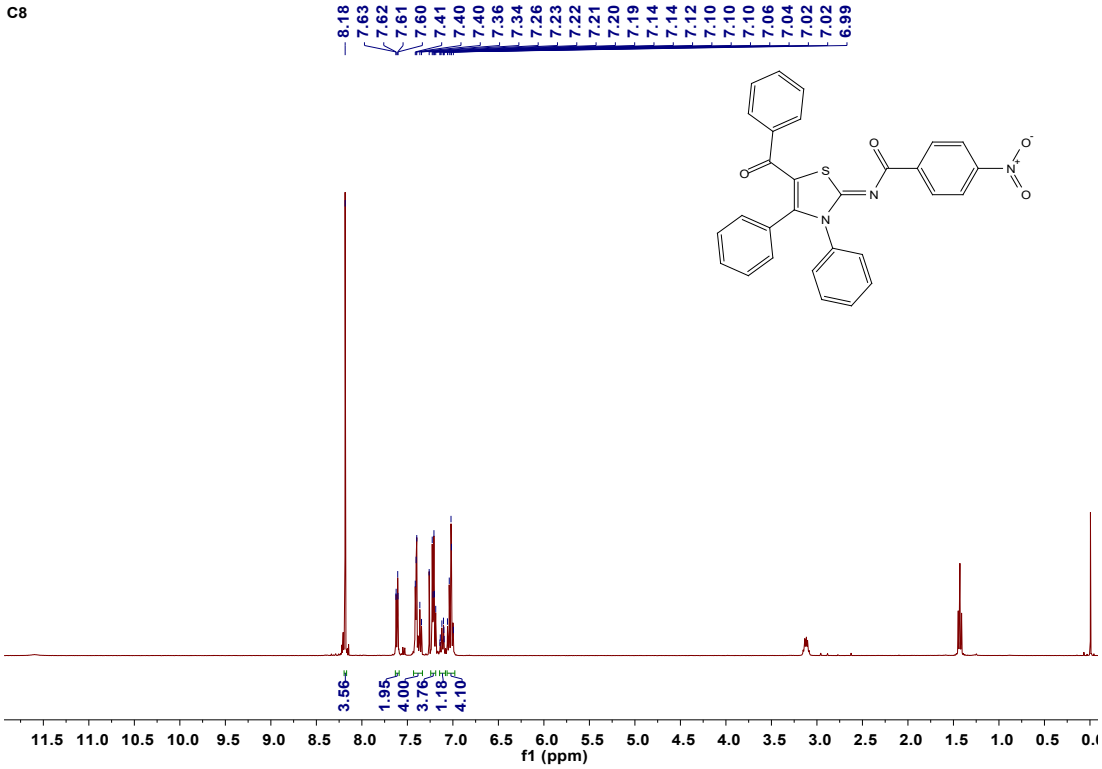


Figure S18 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-nitrobenzamide (c8)

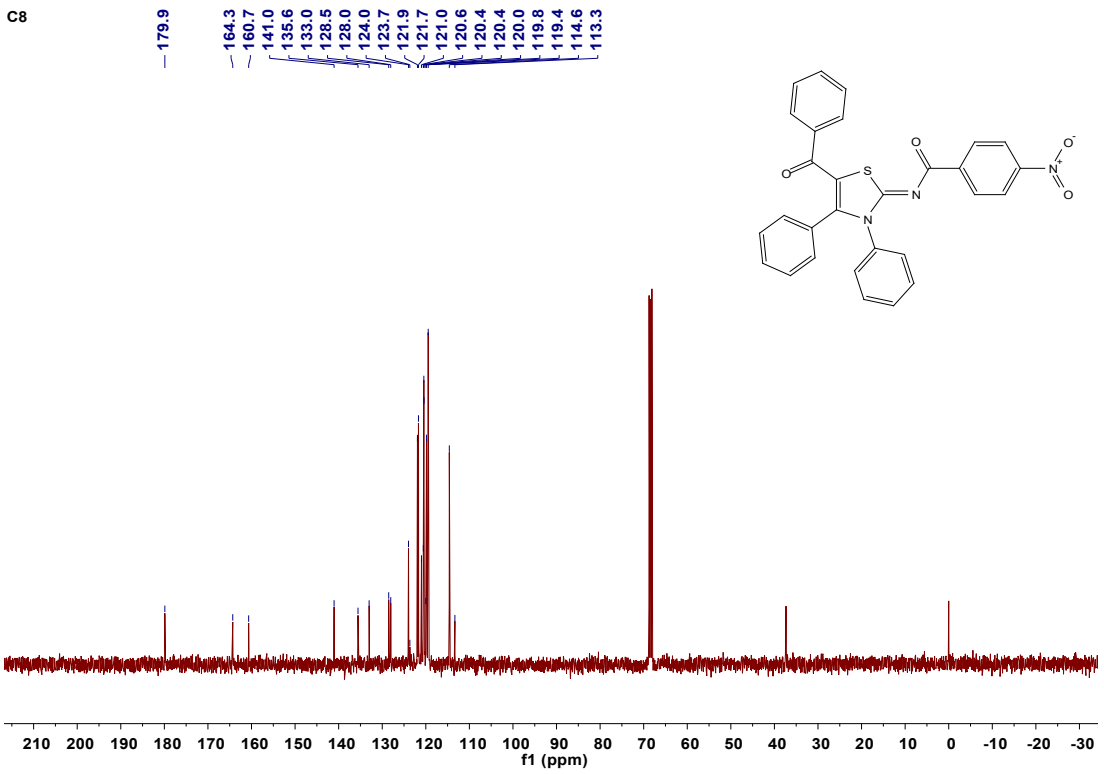


Figure S19 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-4-nitrobenzamide (c8)

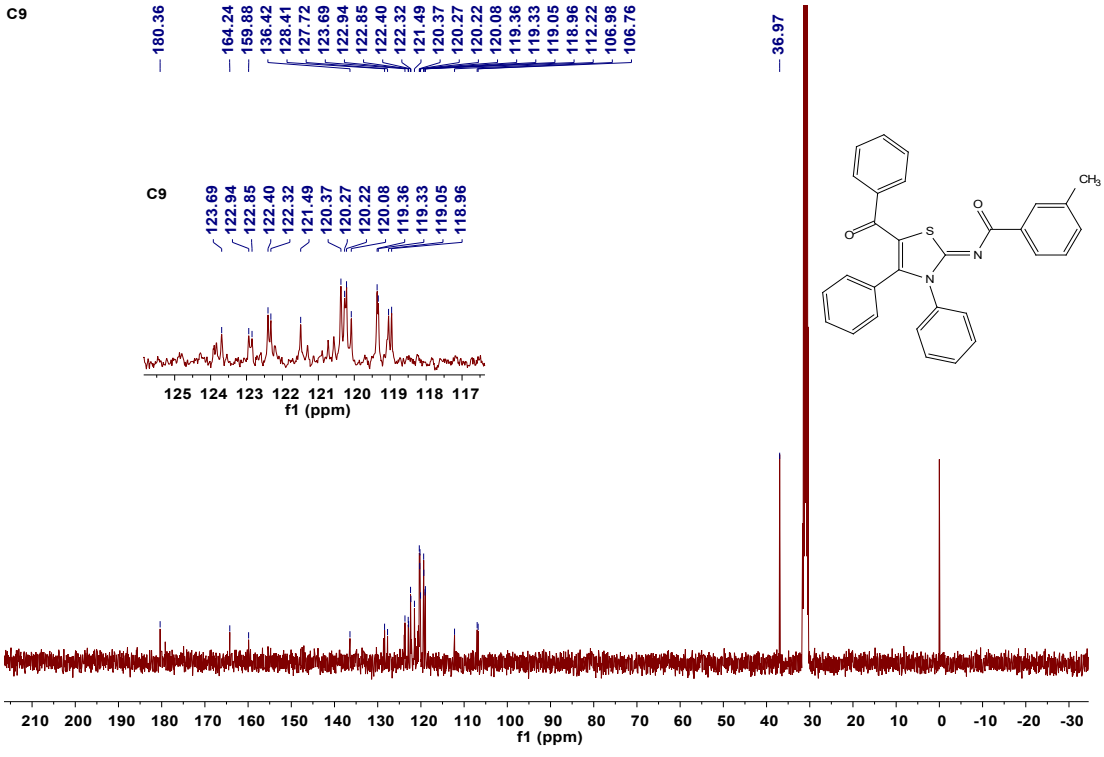


Figure S20 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-3-methylbenzamide (c9)

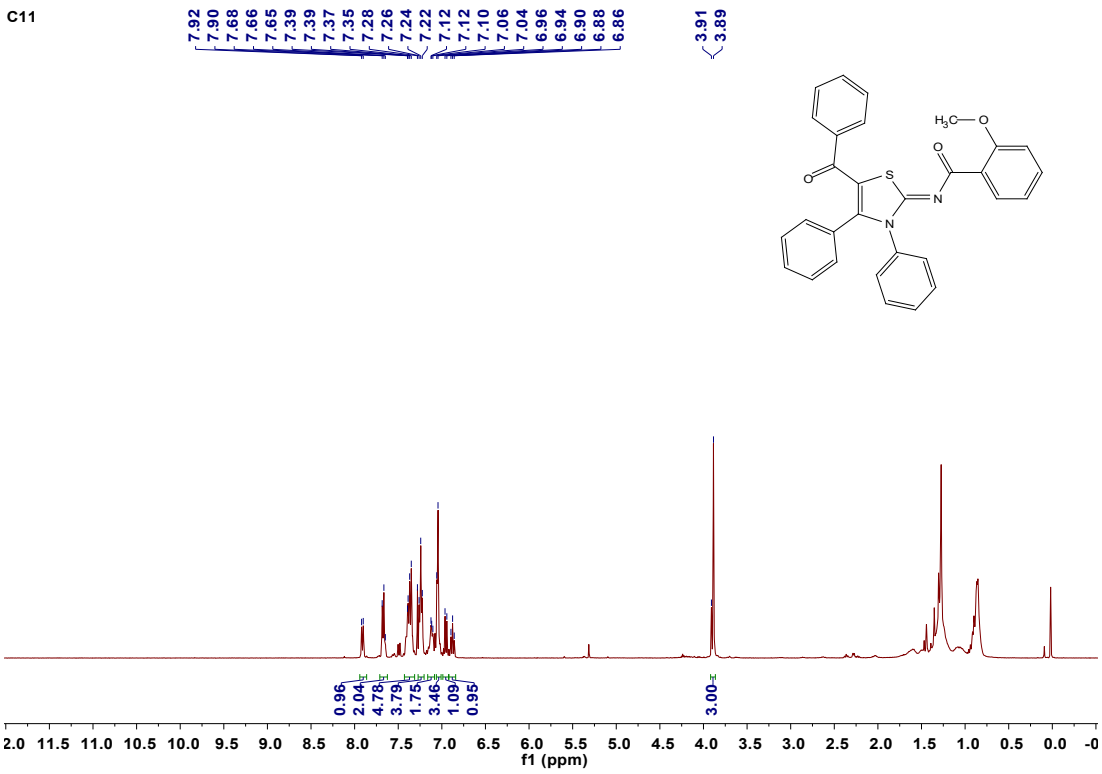


Figure S21 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-methoxybenzamide (c11)

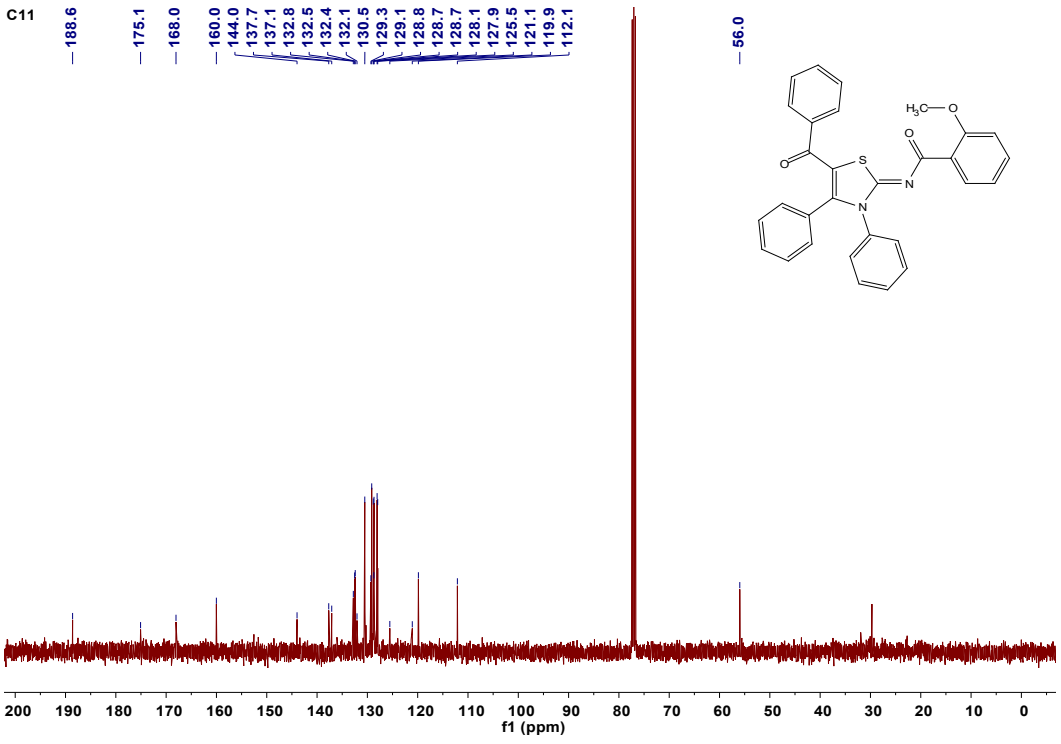


Figure S22 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-methoxybenzamide (c11)

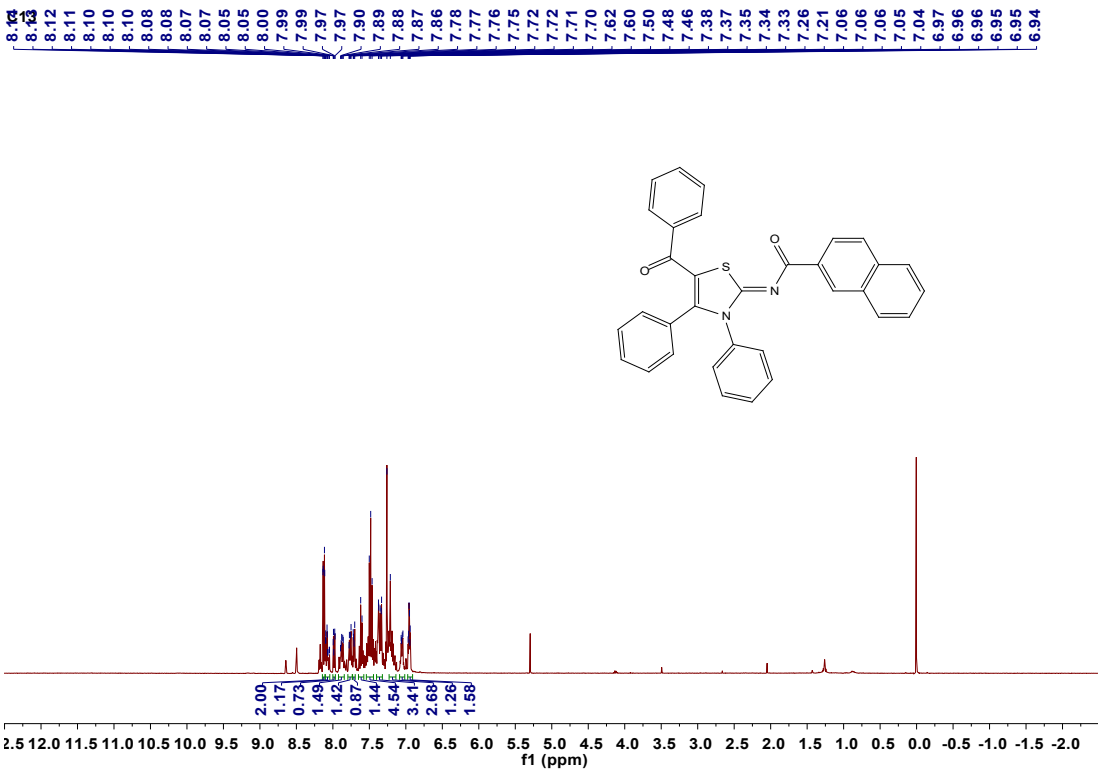


Figure S23 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-naphthamide (c13)

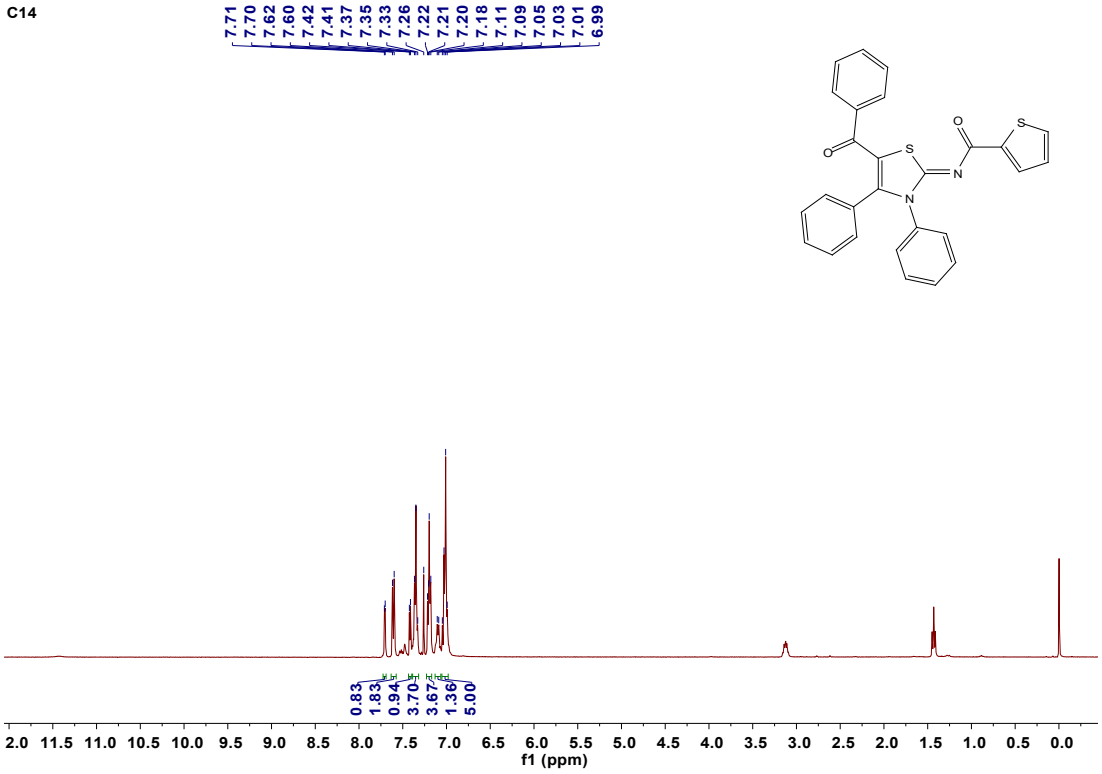


Figure S24 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)thiophene-2-carboxamide (c14)

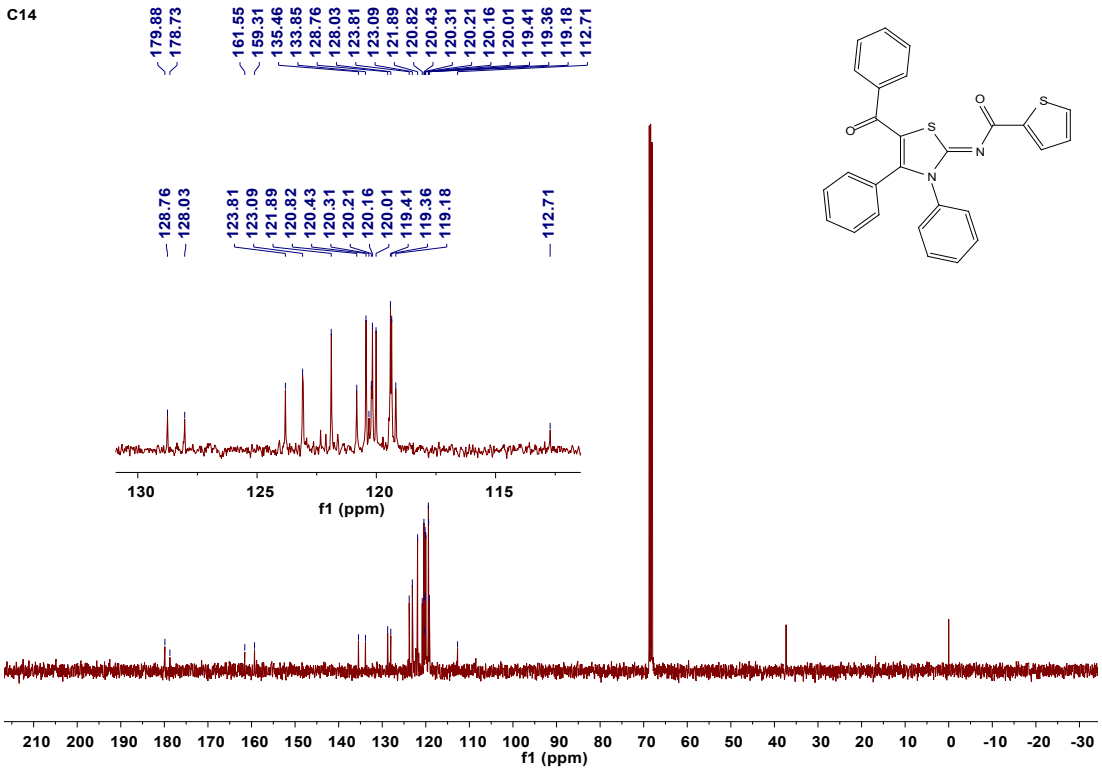


Figure S25 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)thiophene-2-carboxamide (c14)

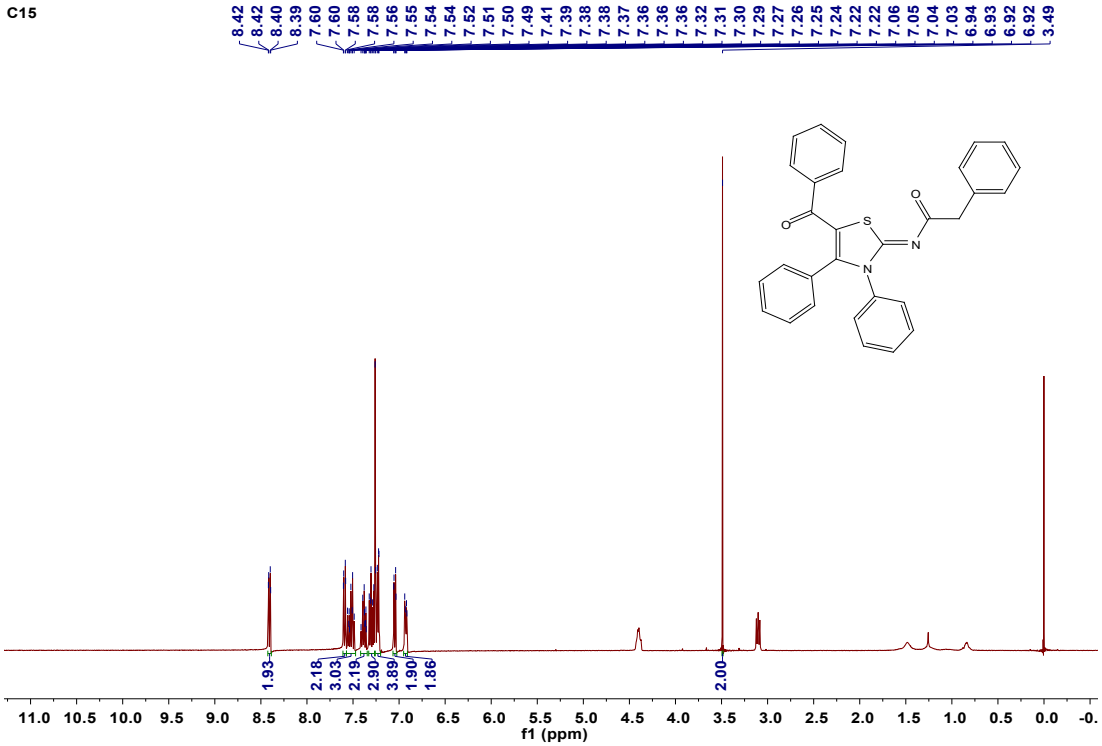


Figure S26 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)-2-phenylacetamide (c15)

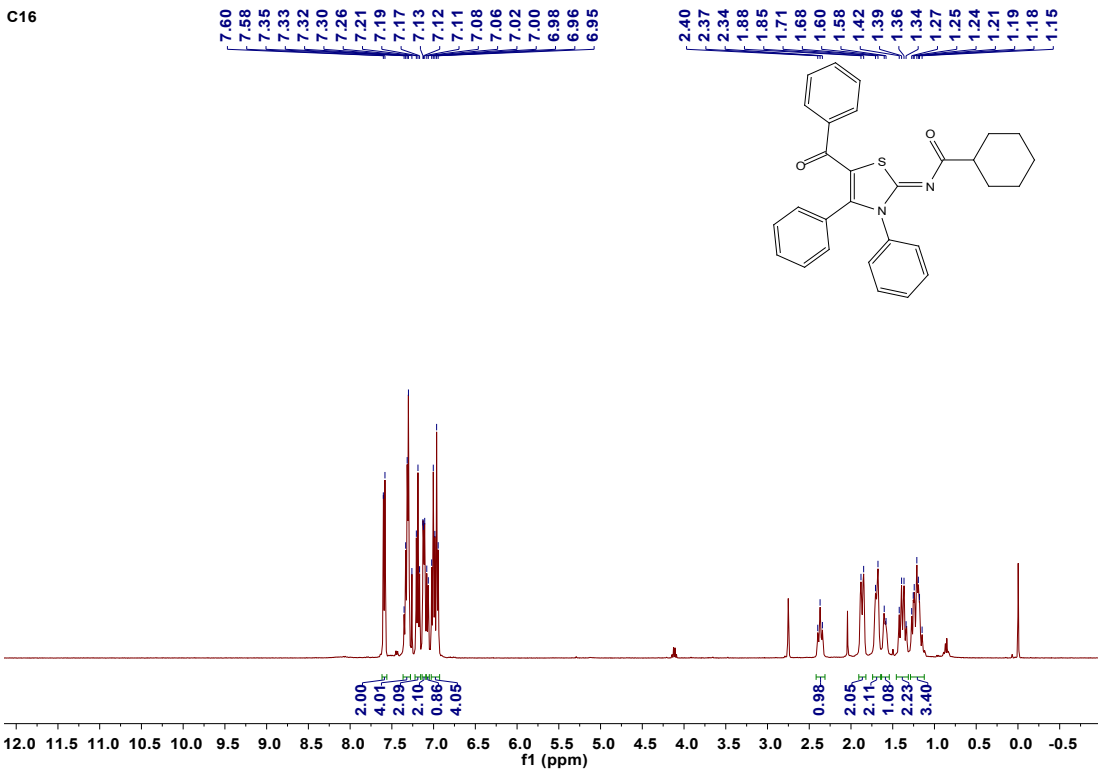


Figure S27 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclohexanecarboxamide (c16)

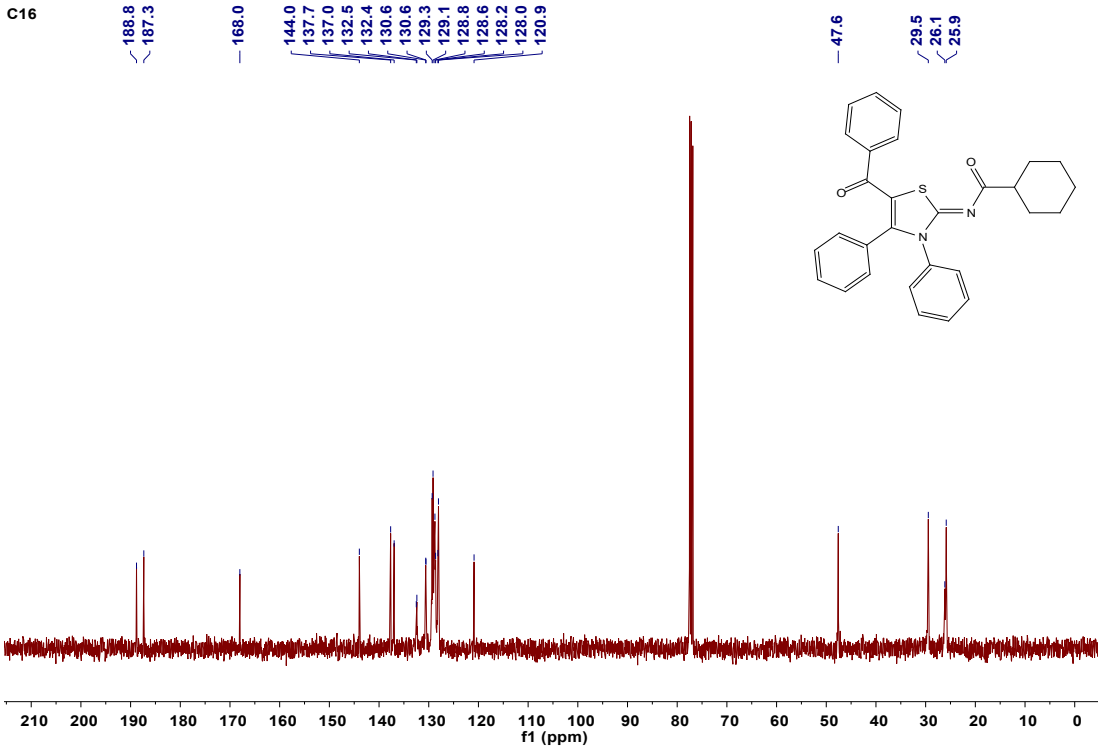


Figure S28 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclohexanecarboxamide (c16)

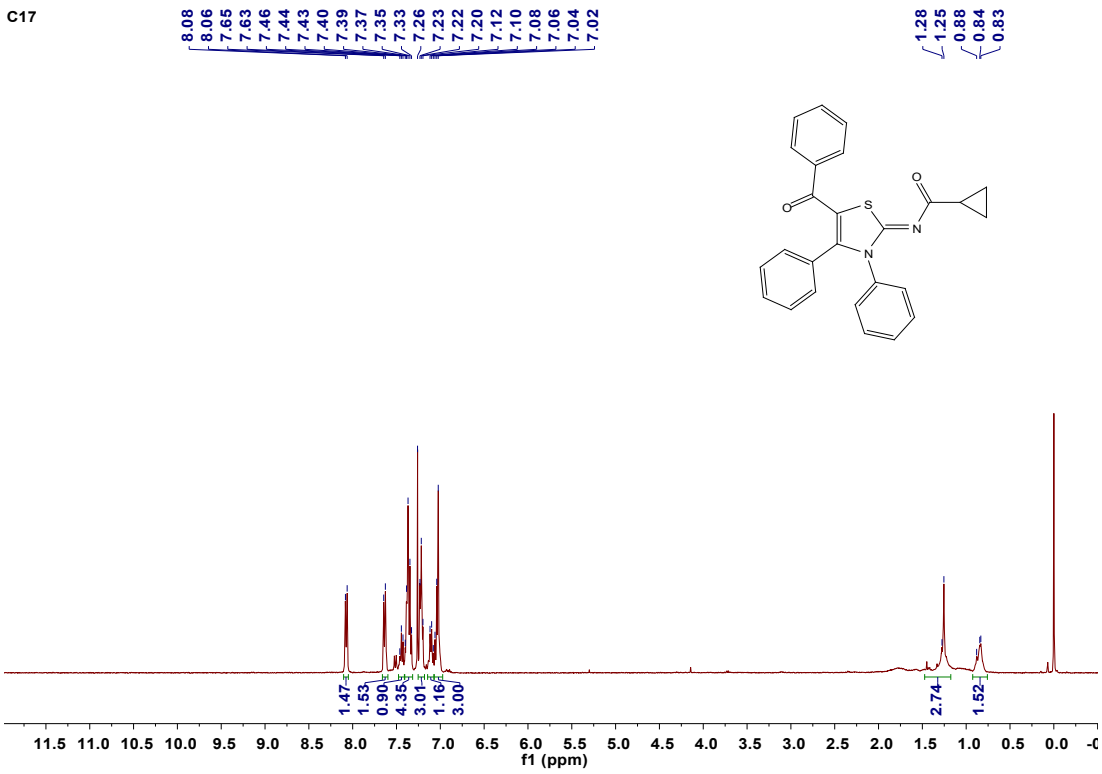


Figure S29 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclopropanecarboxamide (c17)

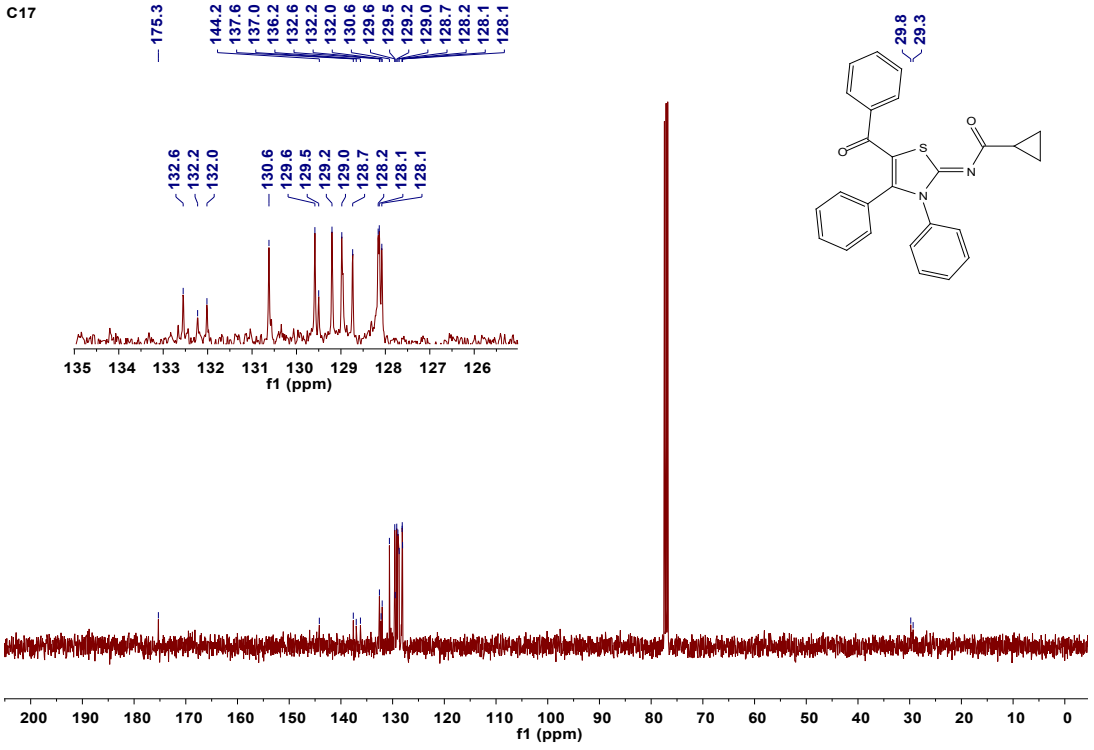


Figure S30 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3,4-diphenylthiazol-2(3H)-ylidene)cyclopropanecarboxamide (c17)

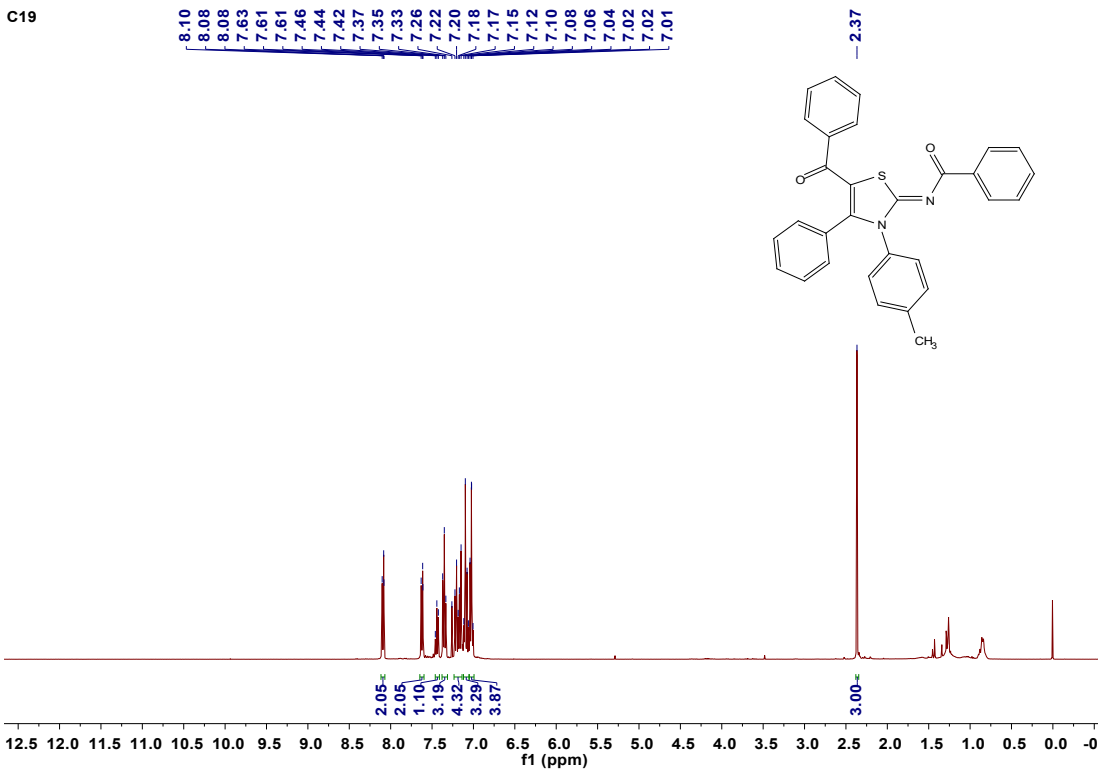


Figure S31 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c19)

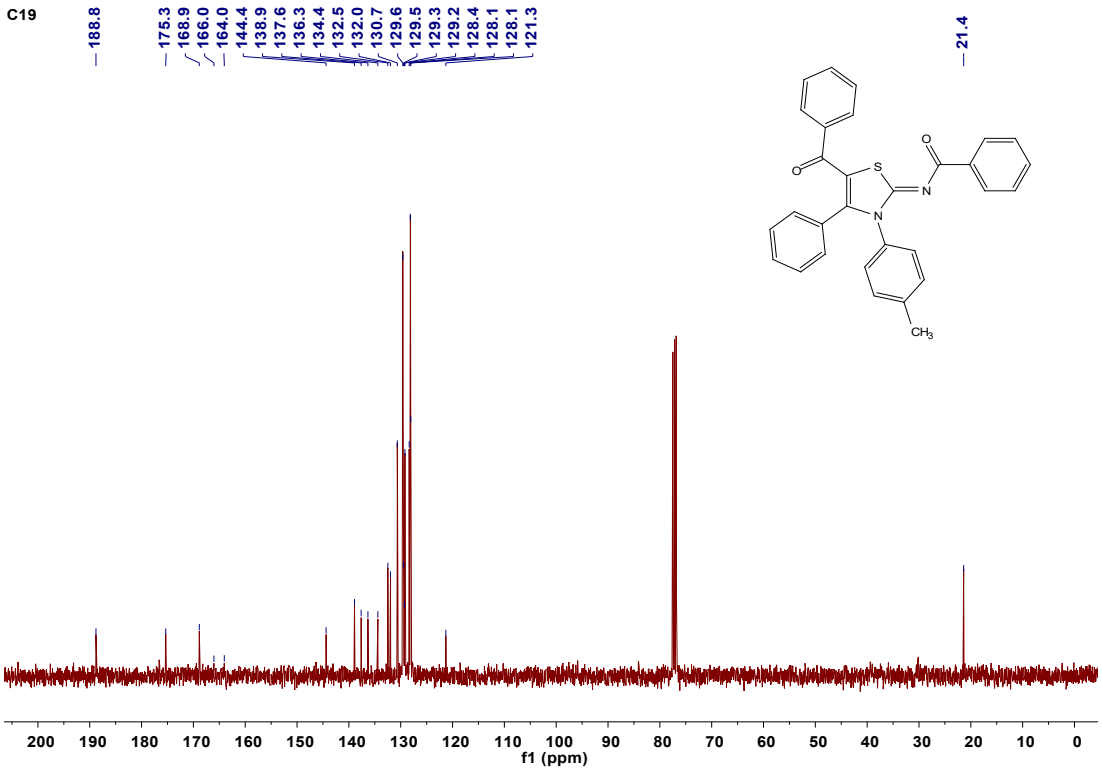


Figure S32 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c19)

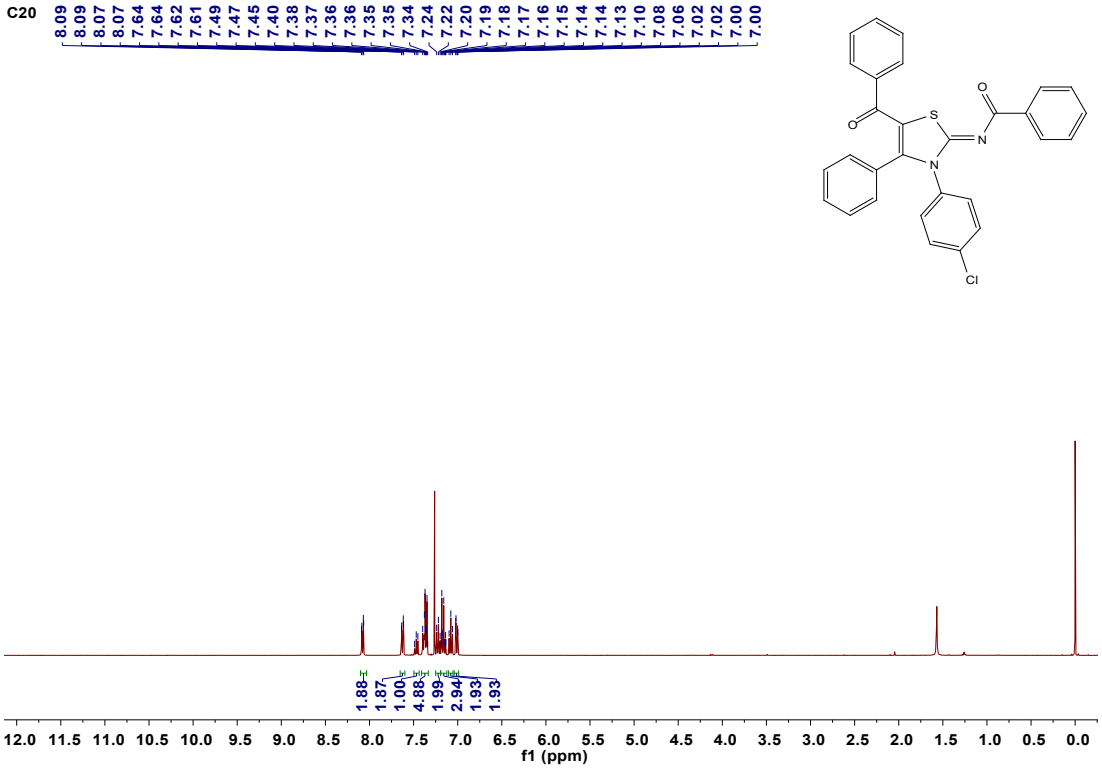


Figure S33 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(4-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c20)

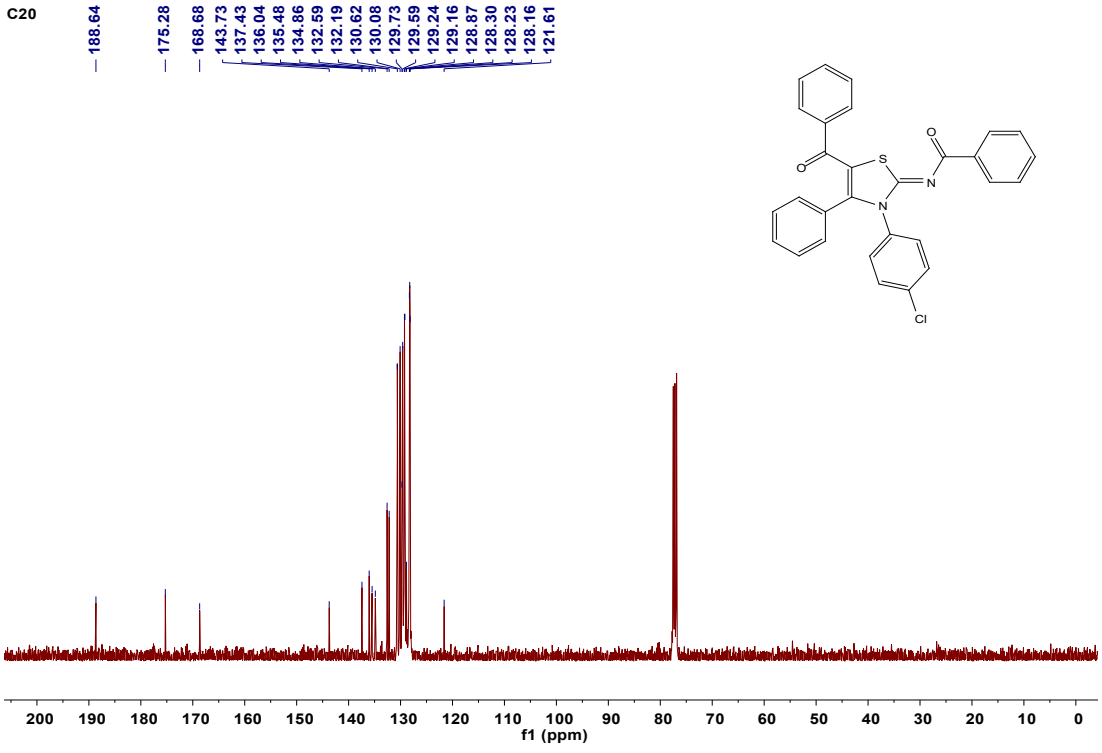


Figure S34 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(4-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c20)

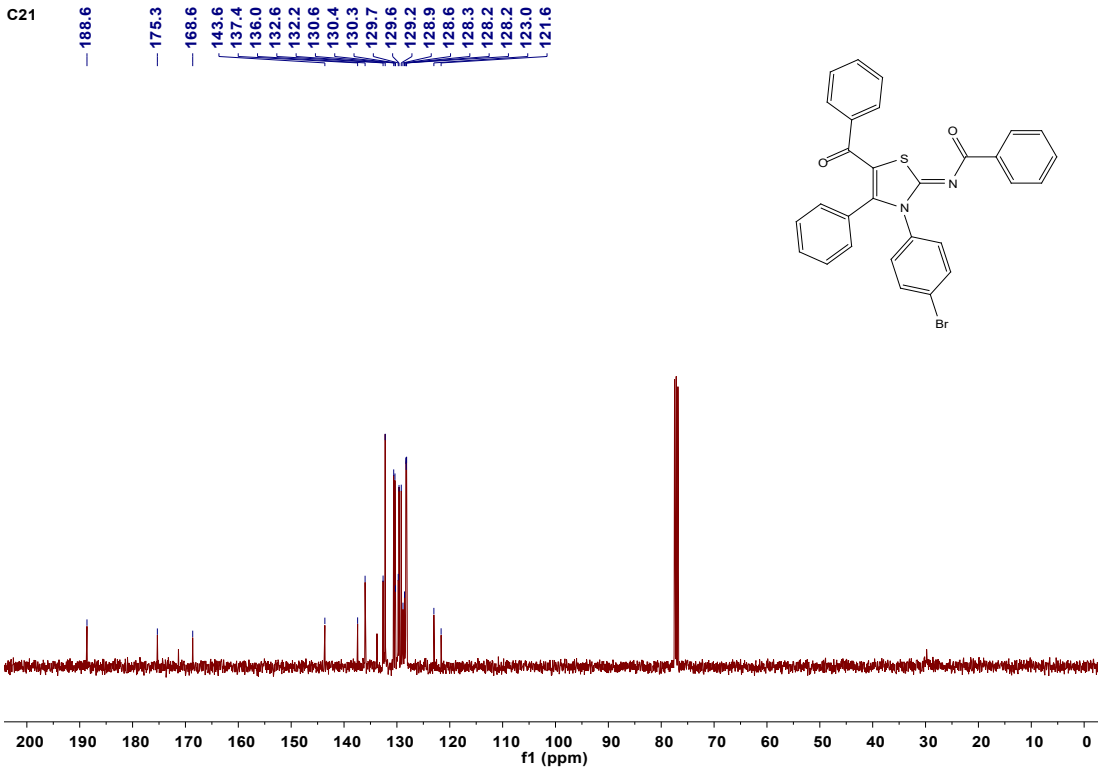


Figure S35 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(4-bromophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c21)

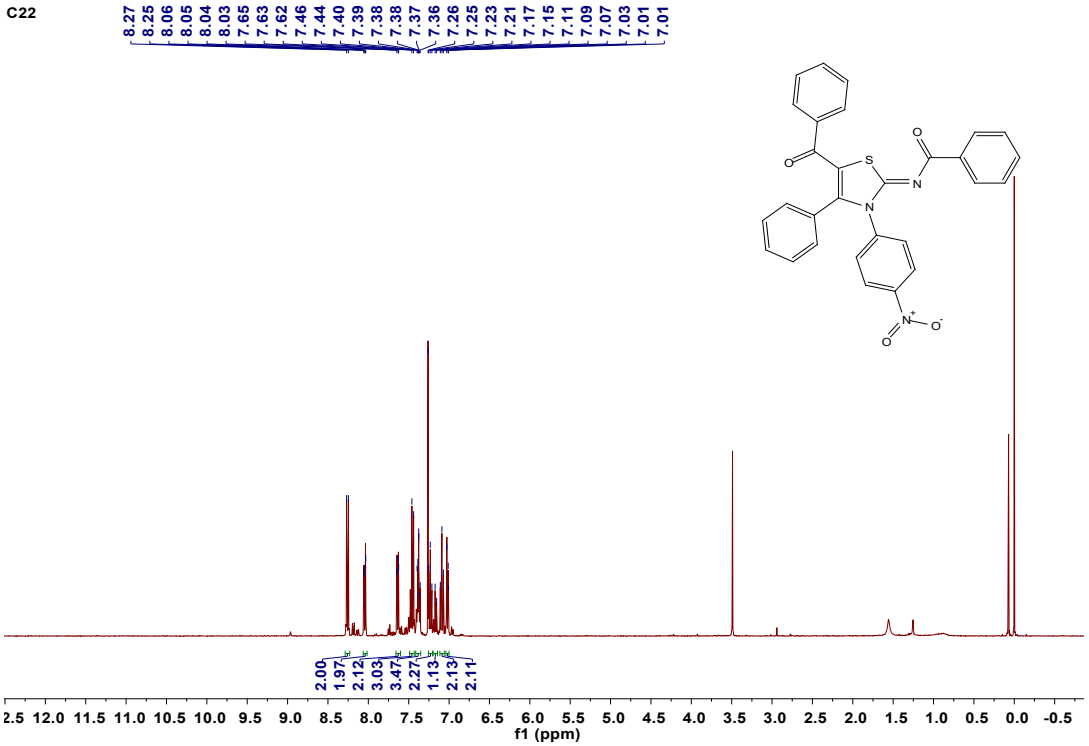


Figure S36 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(4-nitrophenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c22)

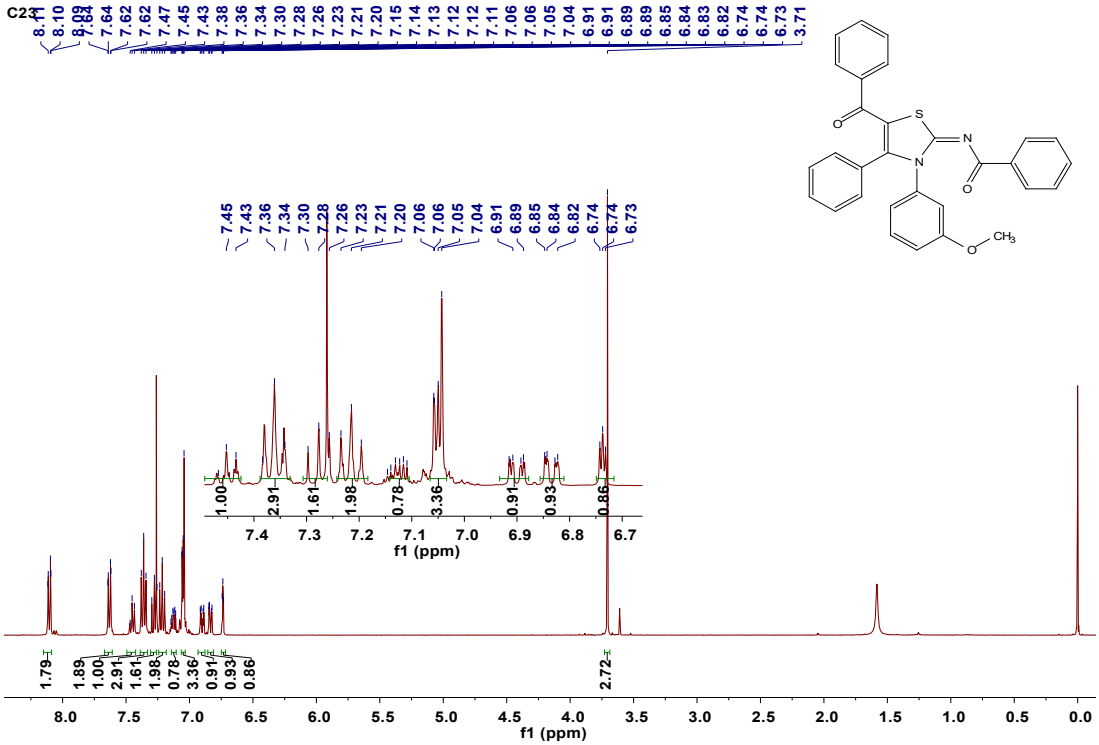


Figure S37 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(3-methoxyphenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c23)

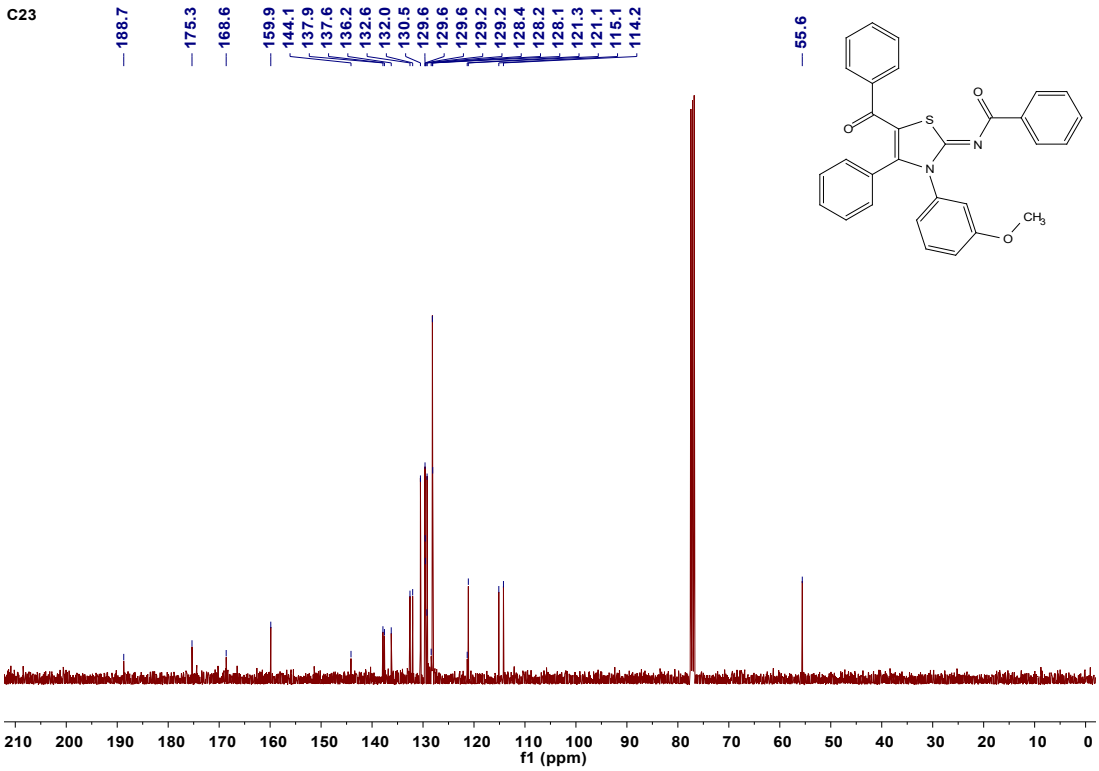


Figure S38 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(3-methoxyphenyl)-4-phenylthiazol-2(3H)-ylidene)benzamide (c23)

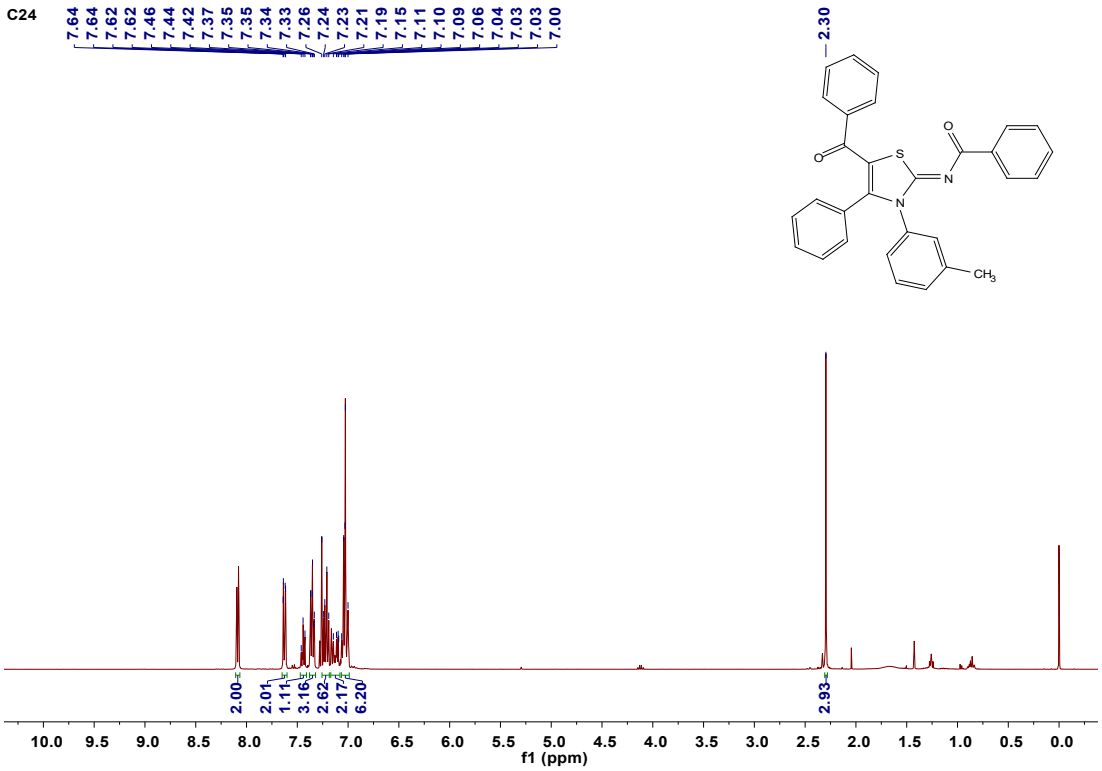


Figure S39 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(m-tolyl)thiazol-2(3H)-ylidene)benzamide (c24)

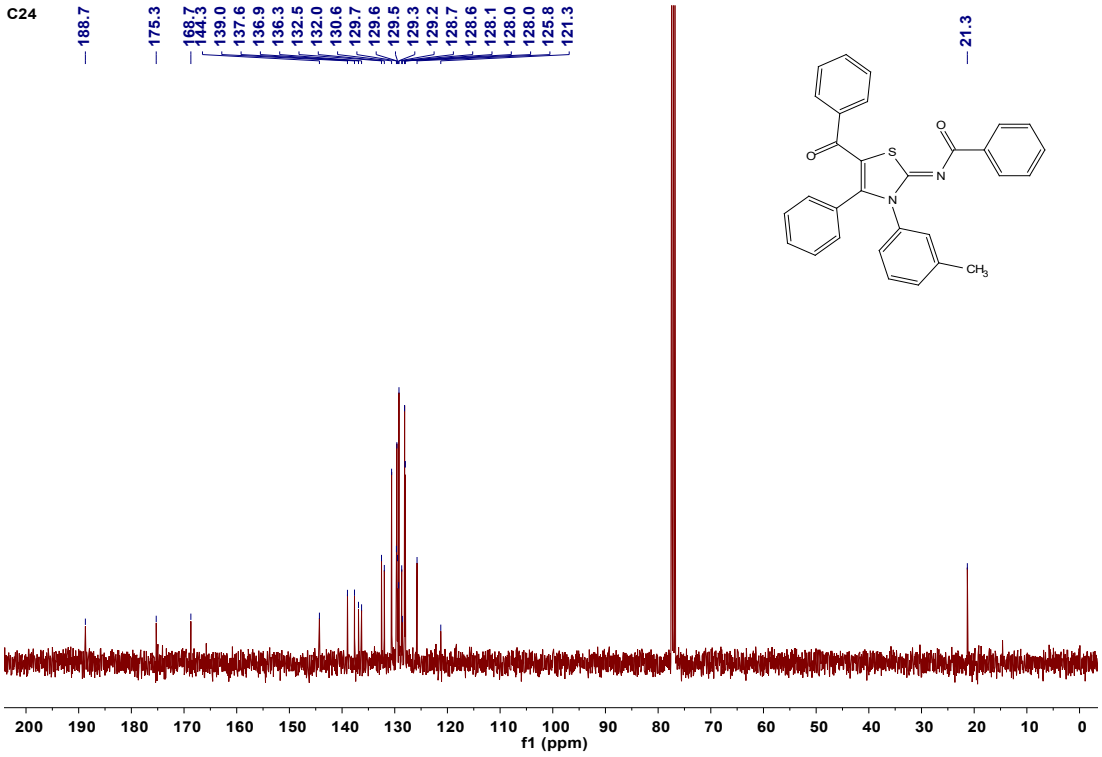


Figure S40 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(m-tolyl)thiazol-2(3H)-ylidene)benzamide (c24)

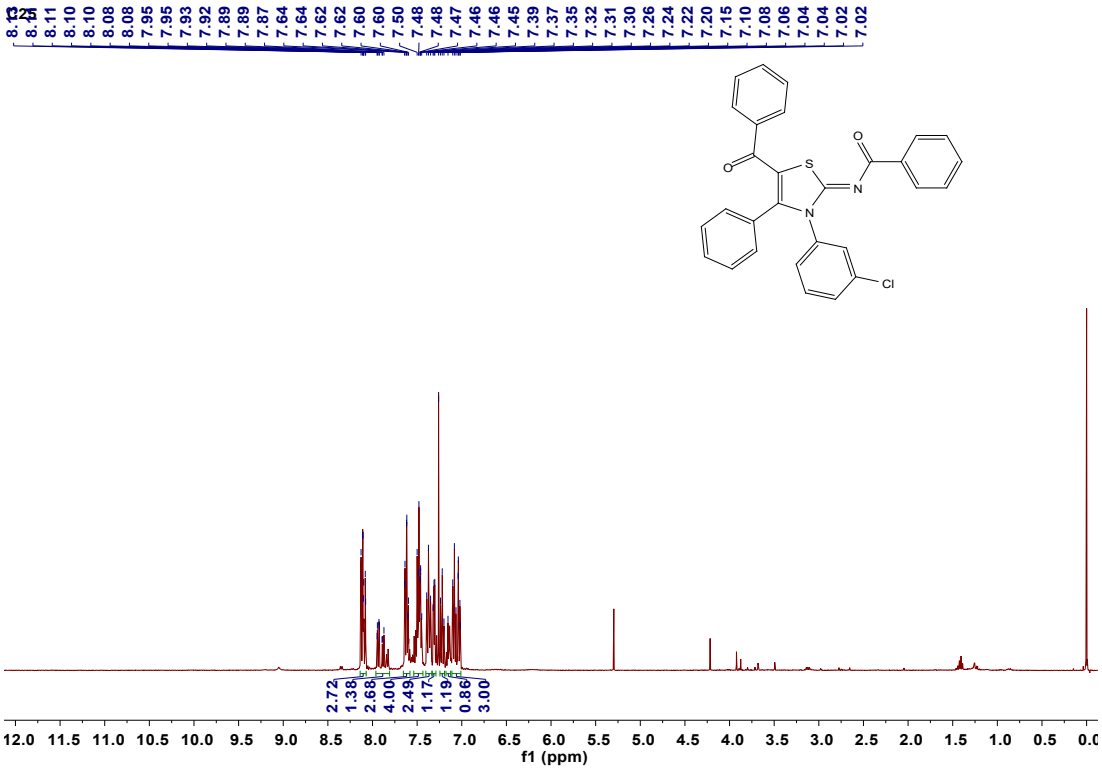


Figure S41 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(3-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c25)

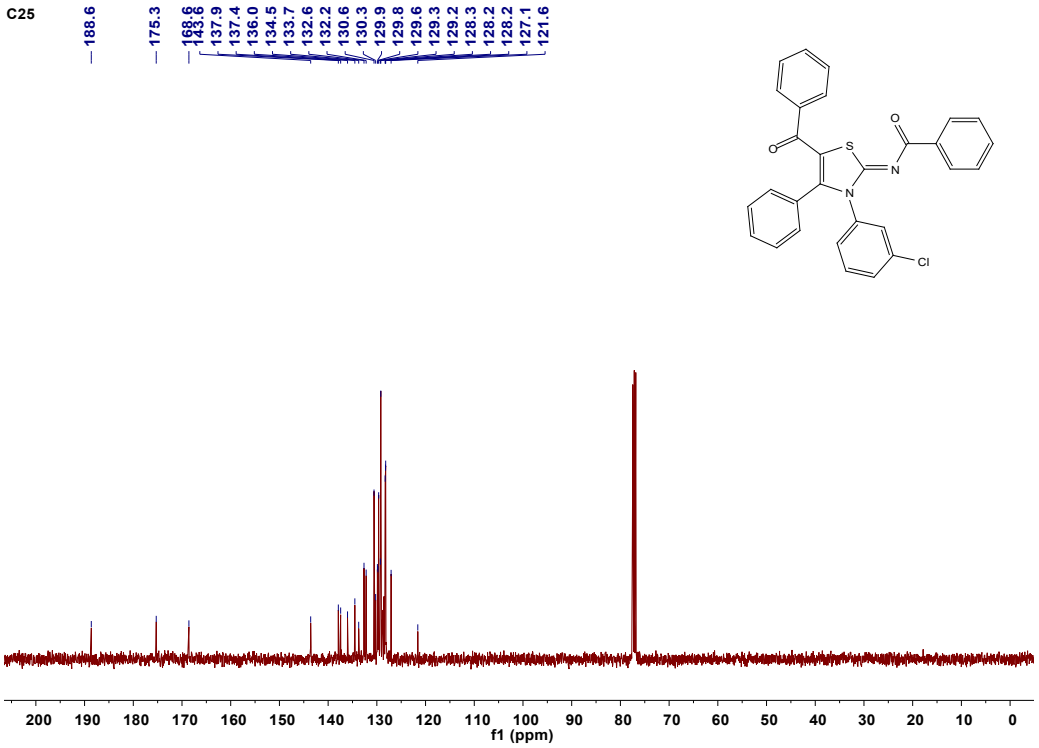


Figure S42 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(3-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c25)

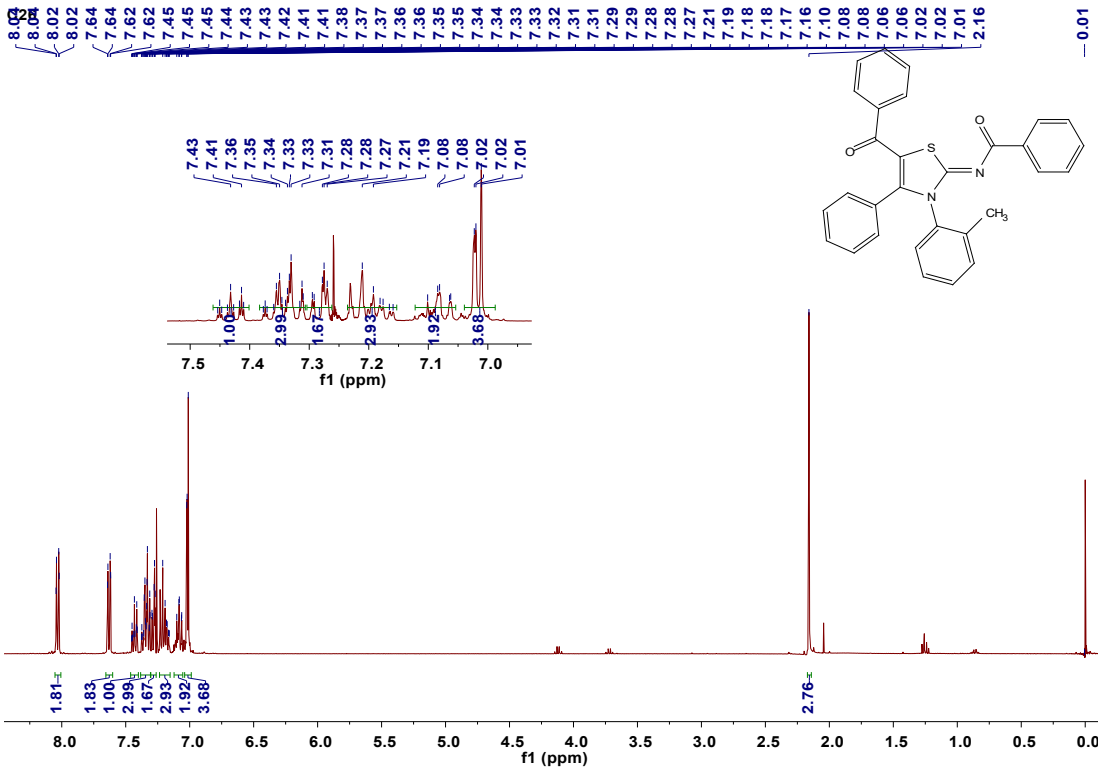


Figure S43 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(o-tolyl)thiazol-2(3H)-ylidene)benzamide (c26)

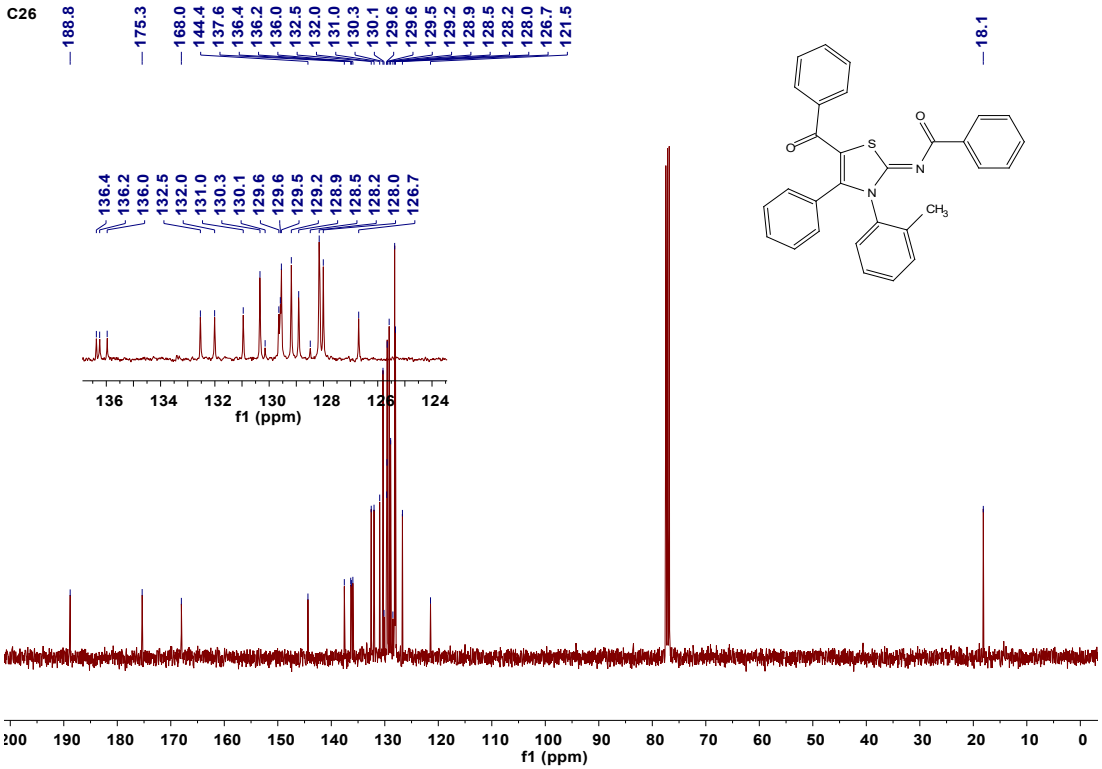


Figure S44 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-phenyl-3-(o-tolyl)thiazol-2(3H)-ylidene)benzamide (c26)

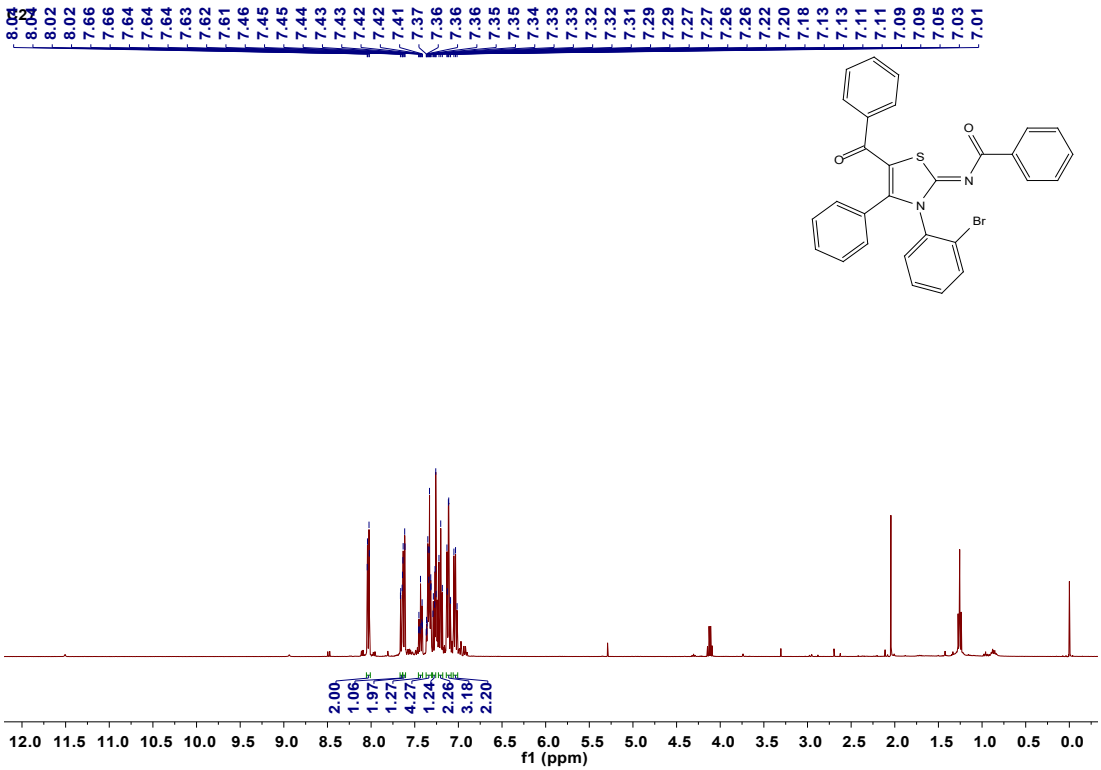


Figure S45 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(2-bromophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c27)

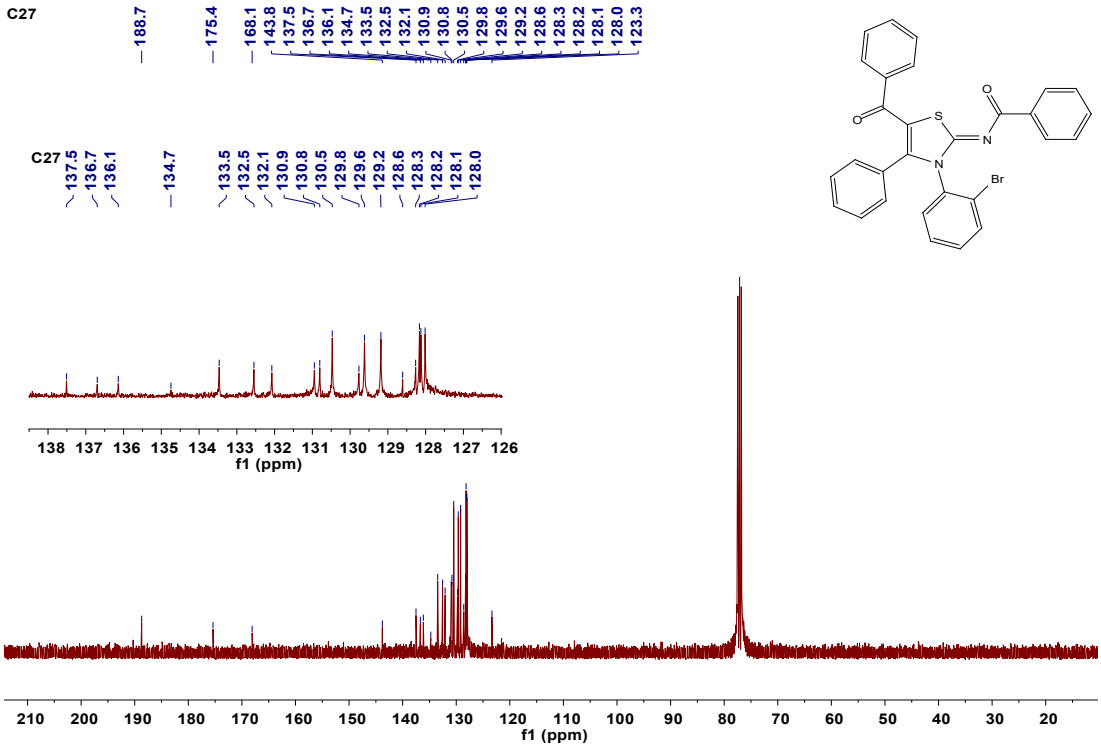


Figure S46 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(2-bromophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c27)

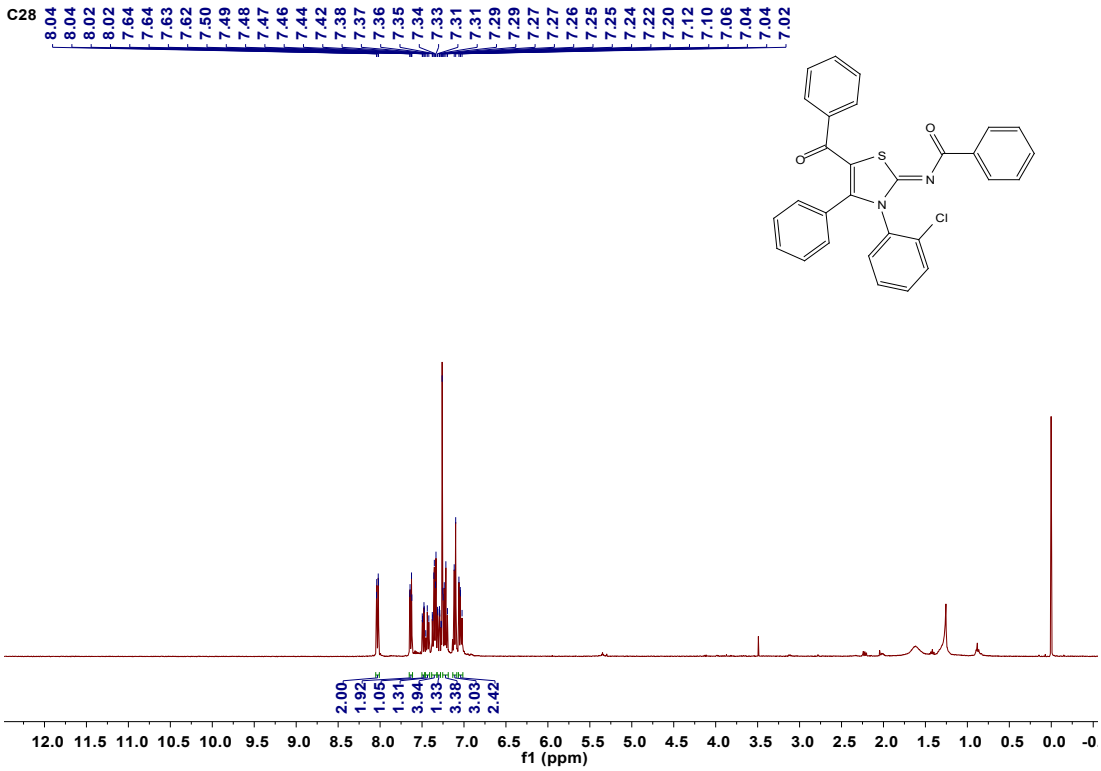


Figure S47 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(2-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c28)

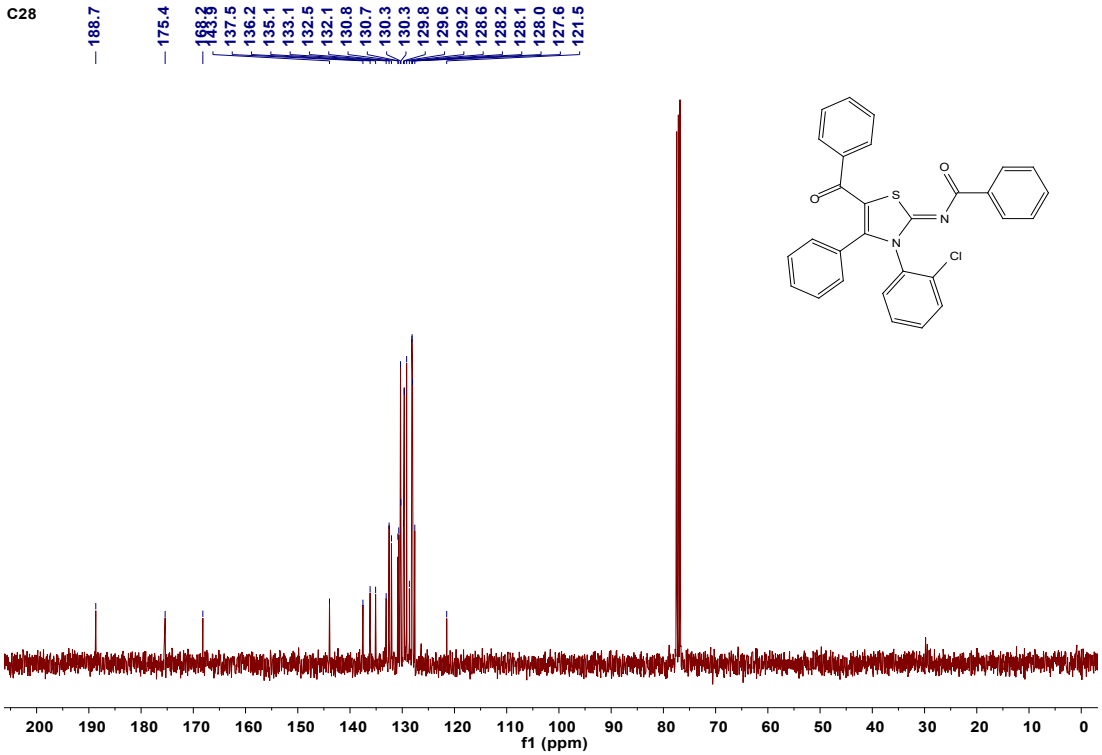


Figure S48 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(2-chlorophenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c28)

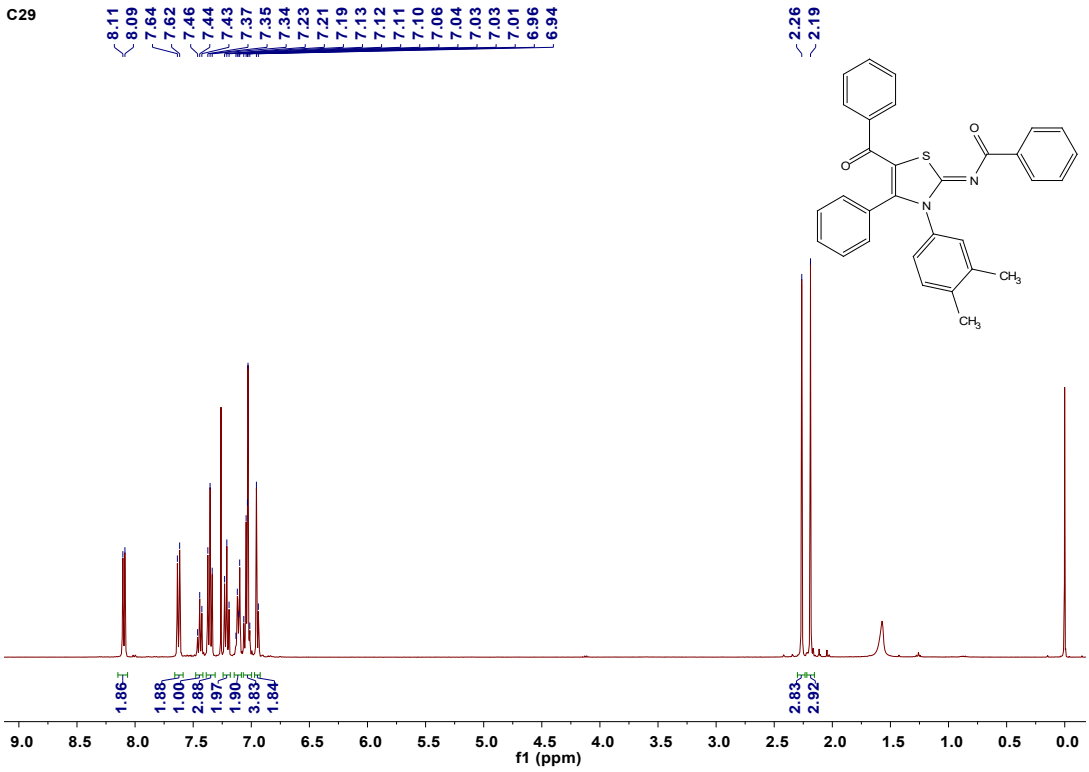


Figure S49 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-(3,4-dimethylphenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c29)

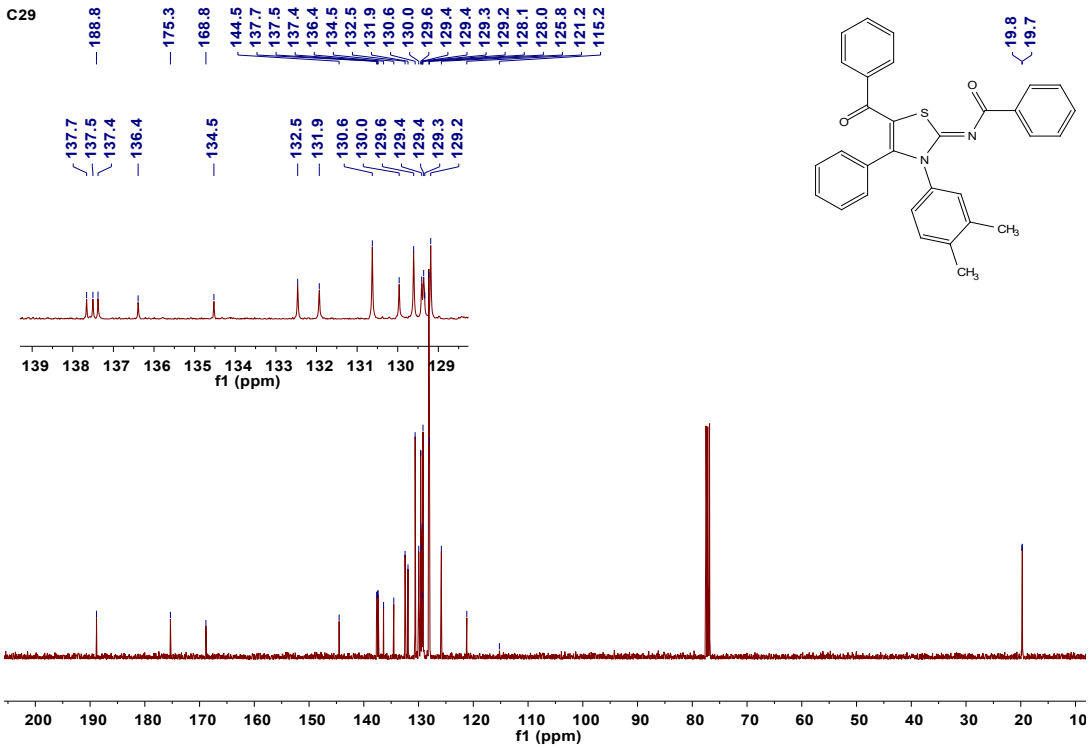


Figure S50 ^{13}C NMR (101 MHz, $CDCl_3$) of N-(5-benzoyl-3-(3,4-dimethylphenyl)-4-phenylthiazol-2(3H)-ylidene) benzamide (c29)

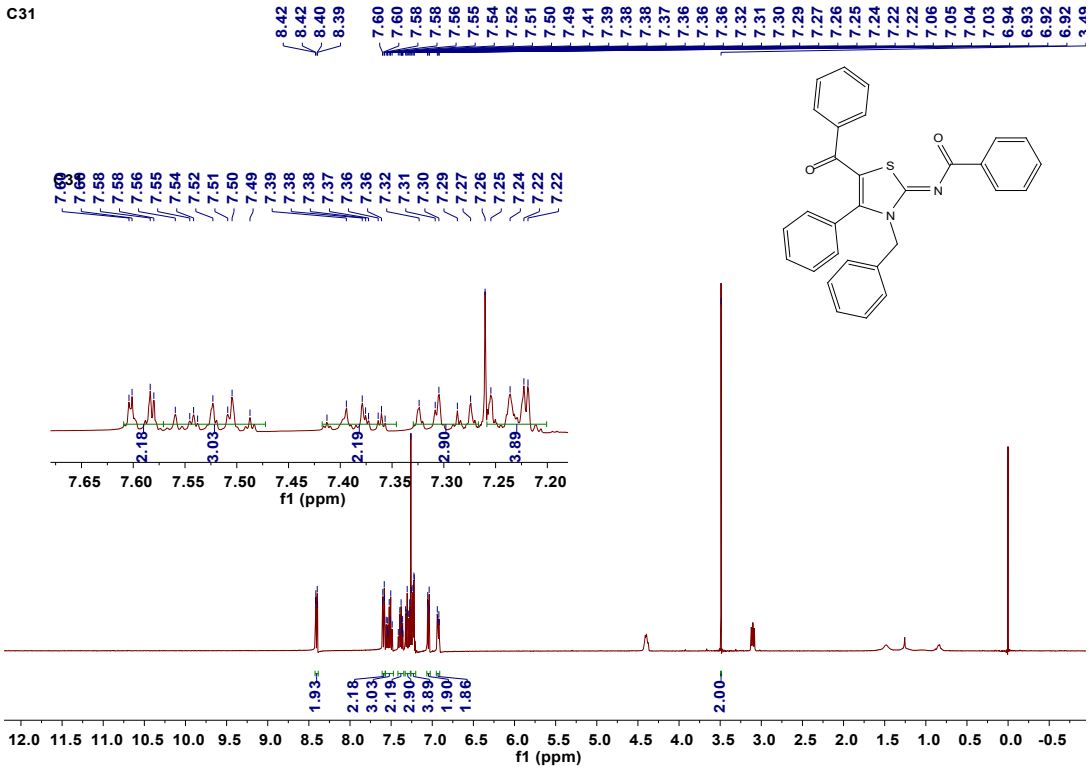


Figure S51 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-benzyl-4-phenylthiazol-2(3H)-ylidene)benzamide (c31)

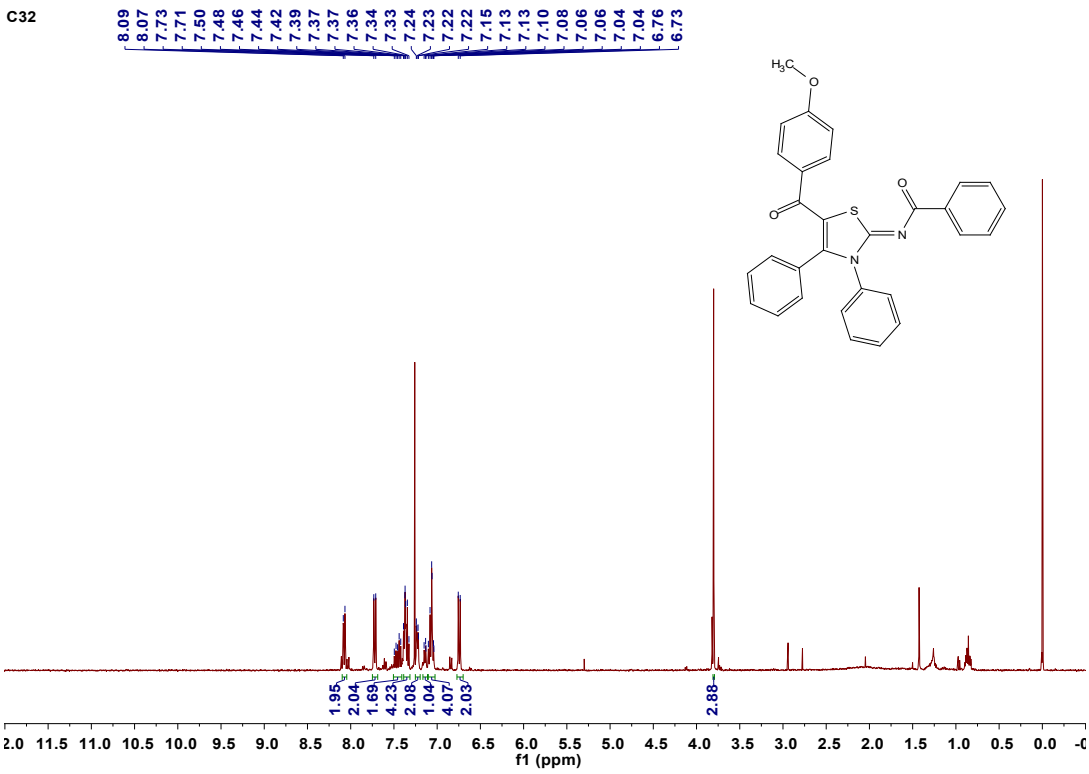


Figure S52 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-(4-methoxybenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c32)

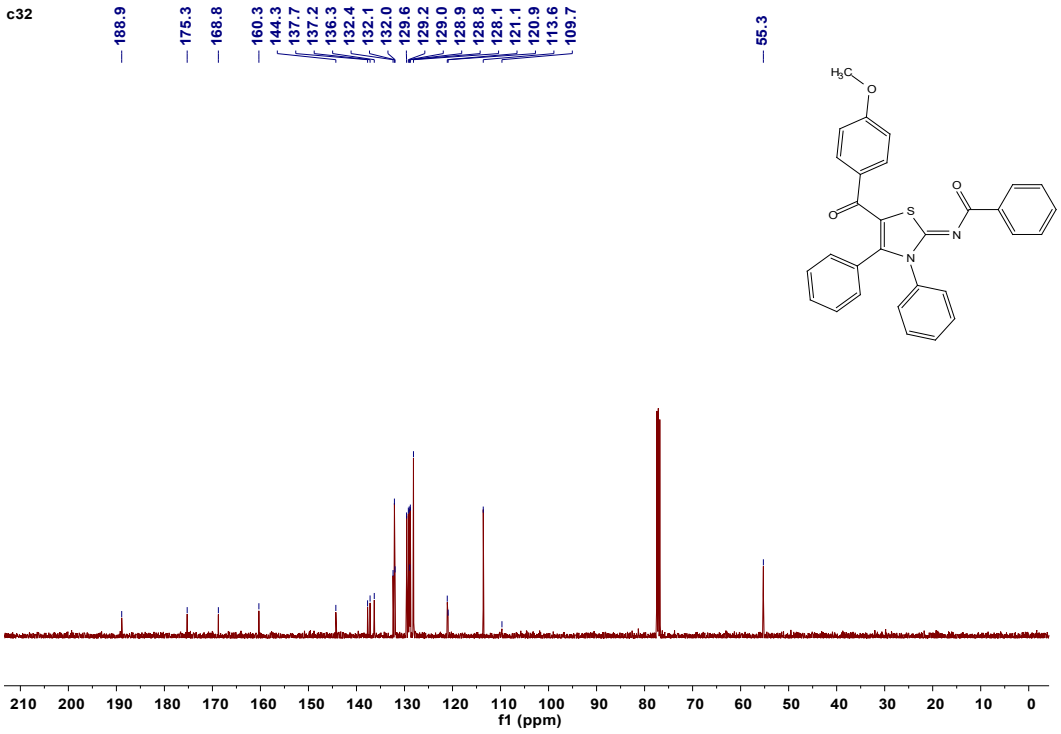


Figure S53 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-(4-methoxybenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene) benzamide (c32)

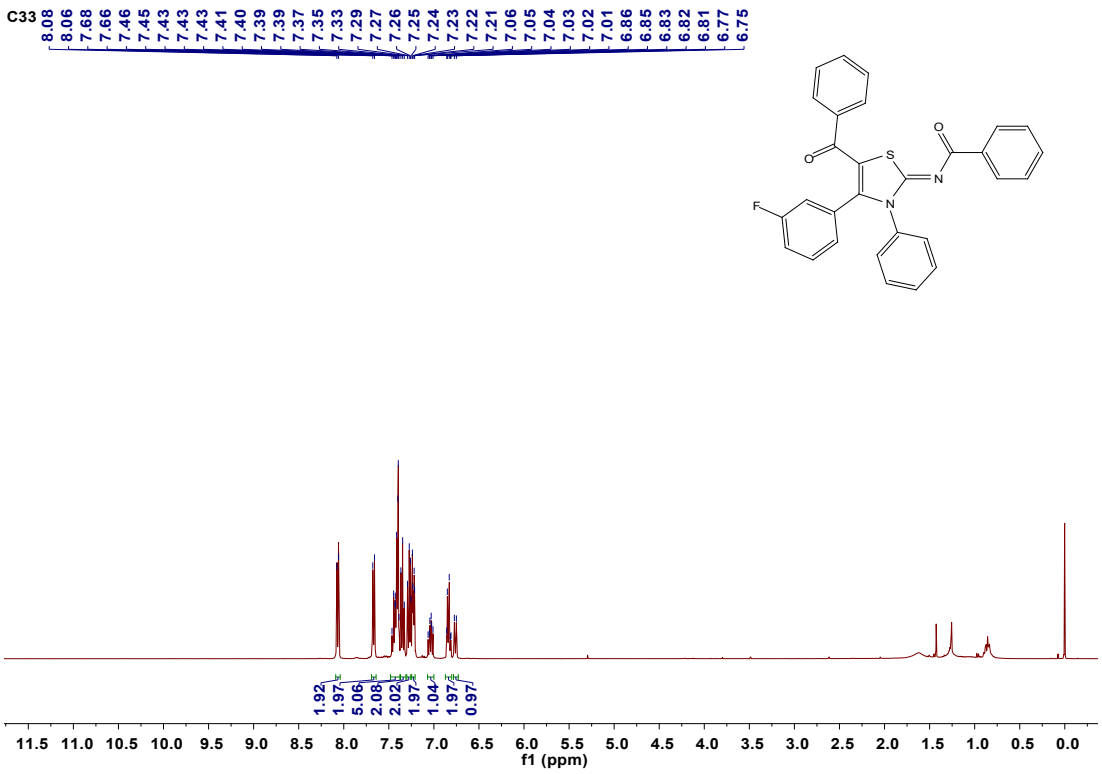


Figure S54 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-(3-fluorophenyl)-3-phenylthiazol-2(3H)-ylidene) benzamide (c33)

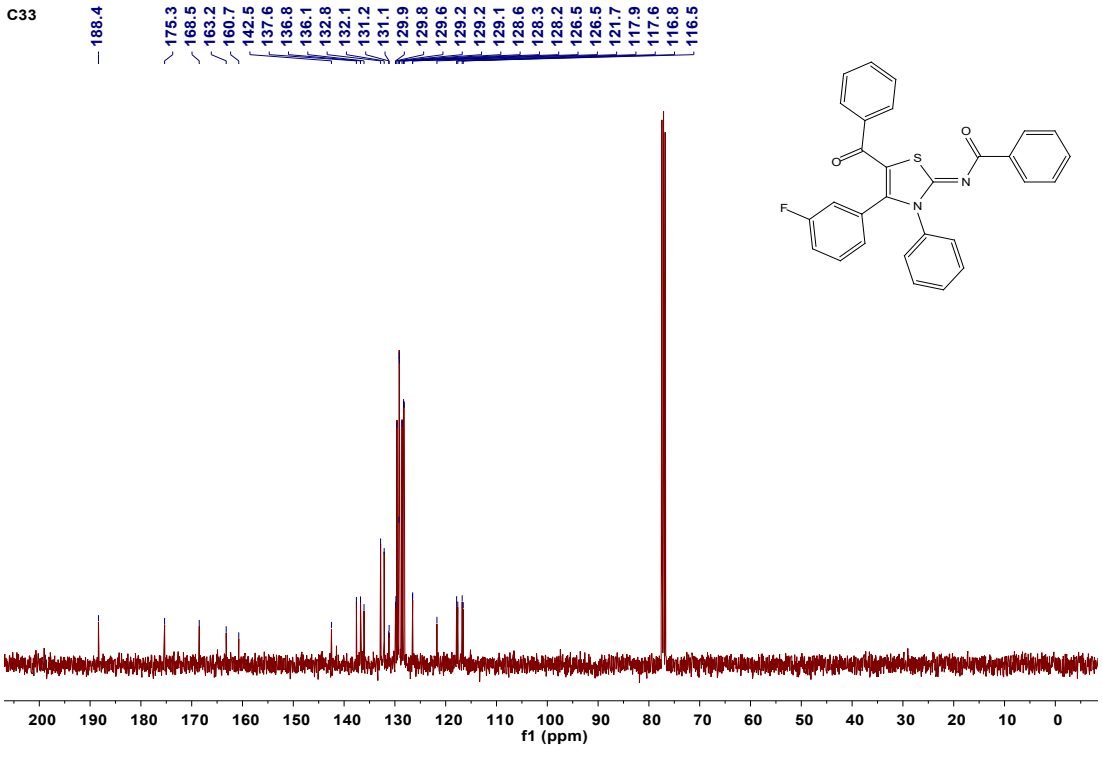


Figure S55 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-4-(3-fluorophenyl)-3-phenylthiazol-2(3H)-ylidene) benzamide (c33)

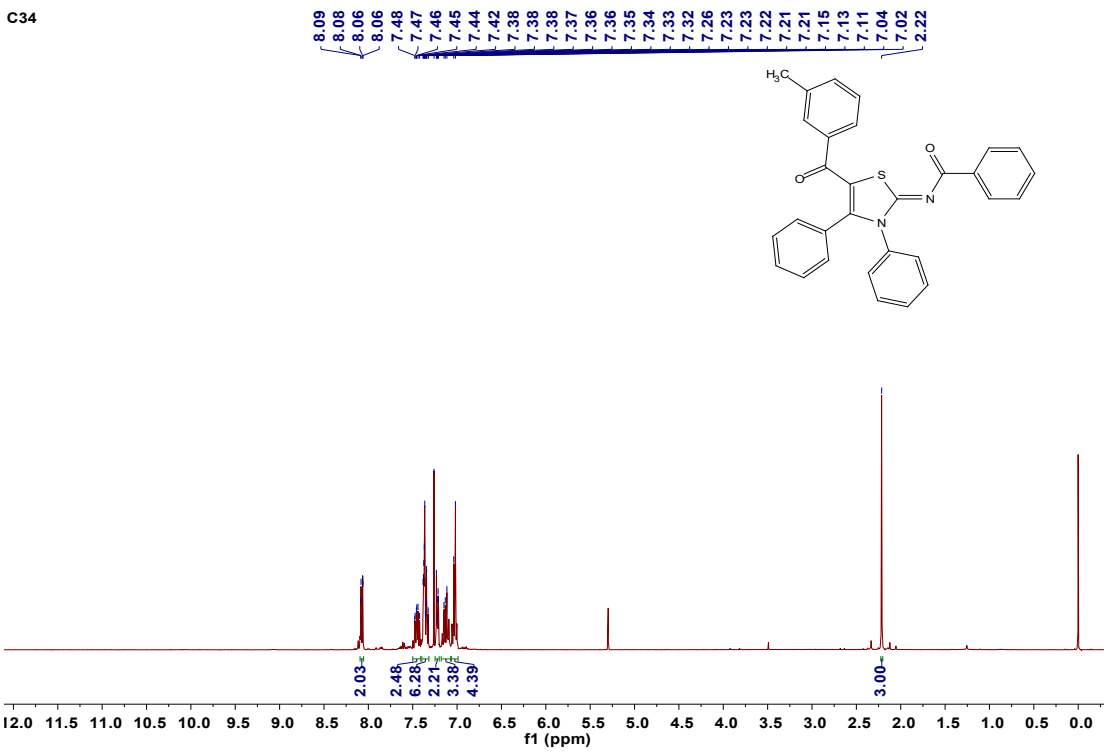


Figure S56 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-(3-methylbenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c34)

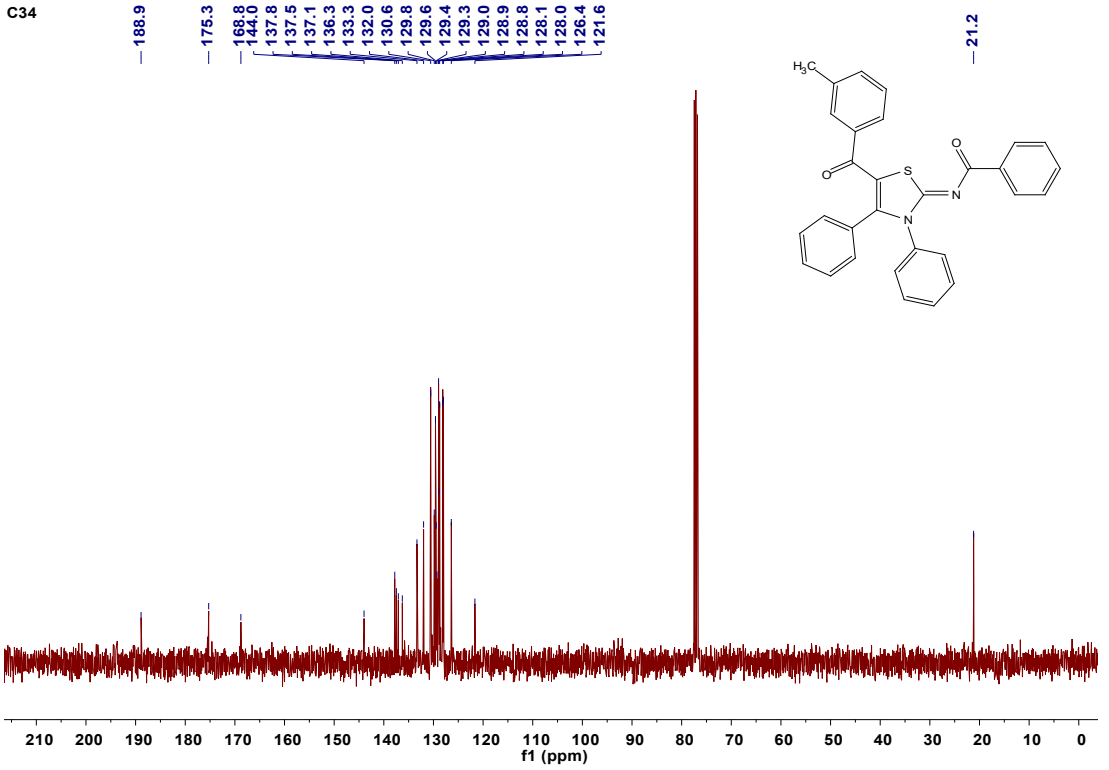


Figure S57 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-(3-methylbenzoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c34)

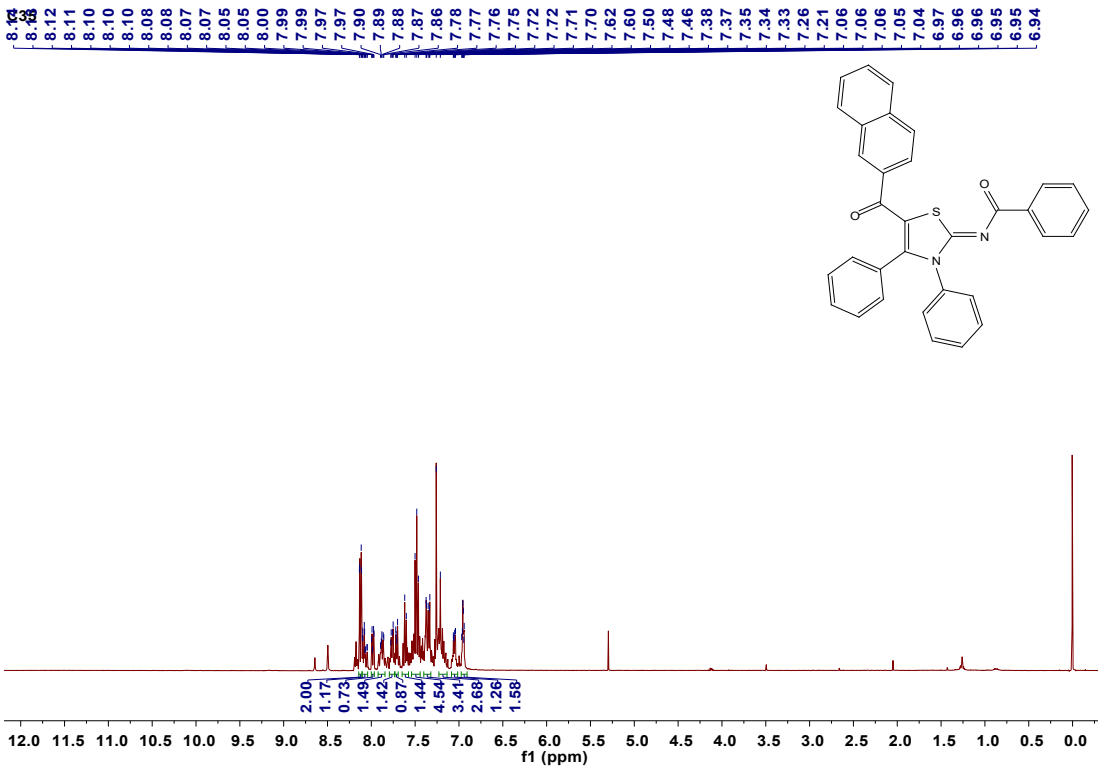


Figure S58 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-(2-naphthoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c35)

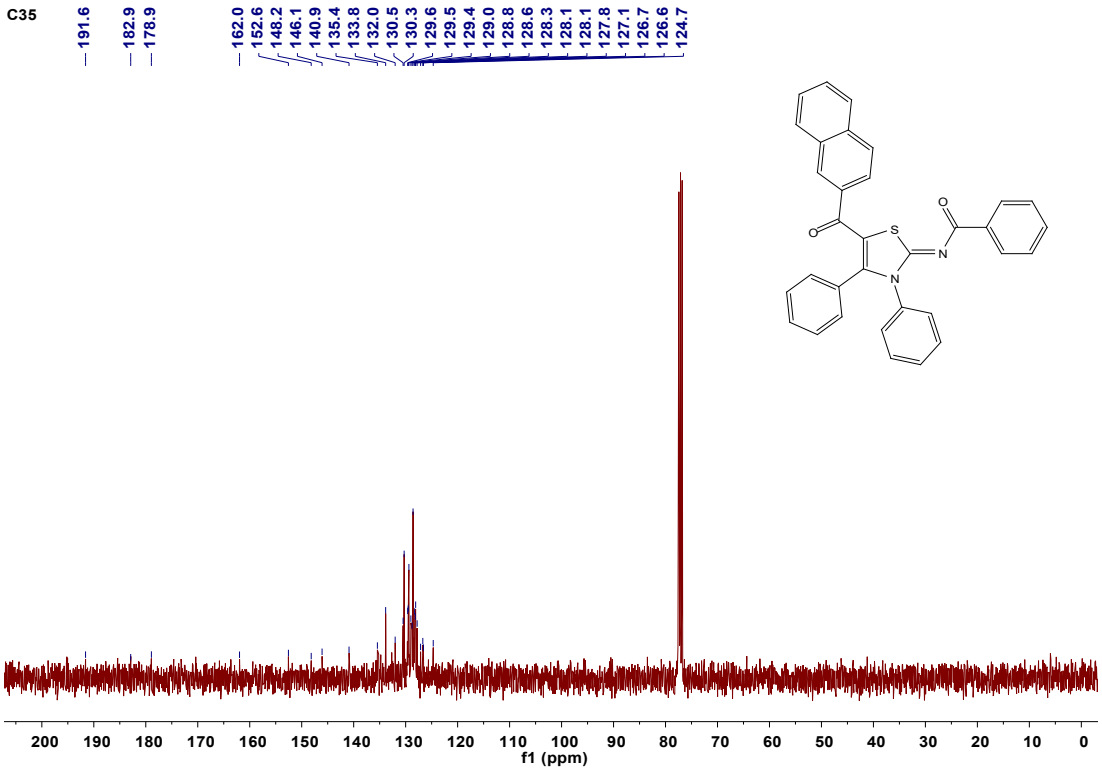


Figure S59 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-(2-naphthoyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c35)

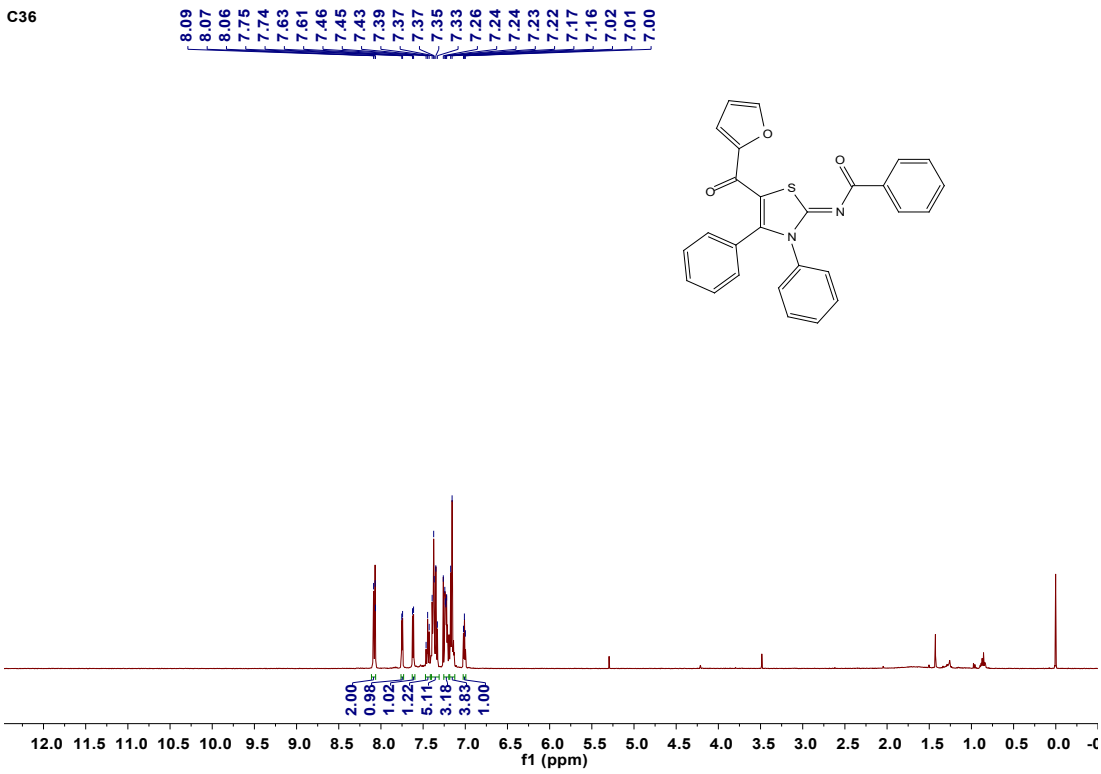


Figure S60 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-(furan-2-carbonyl)-3,4-diphenylthiazol-2(3H)-ylidene)benzamide (c36)

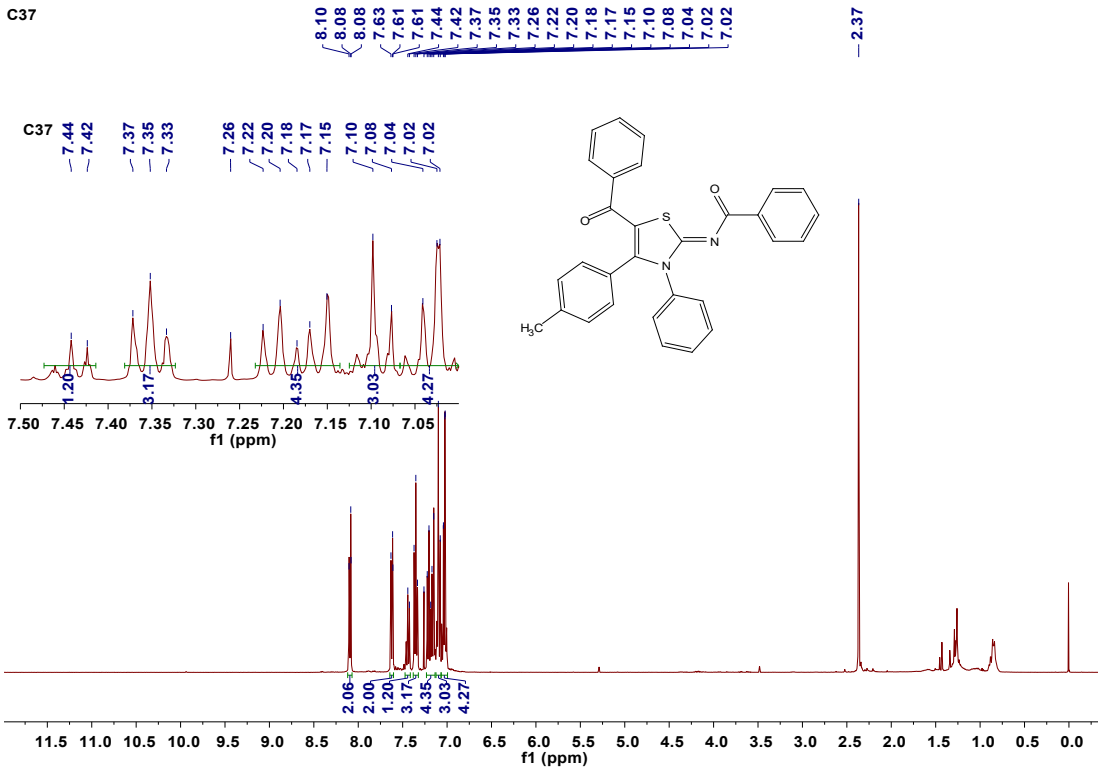


Figure S61 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-phenyl-4-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c37)

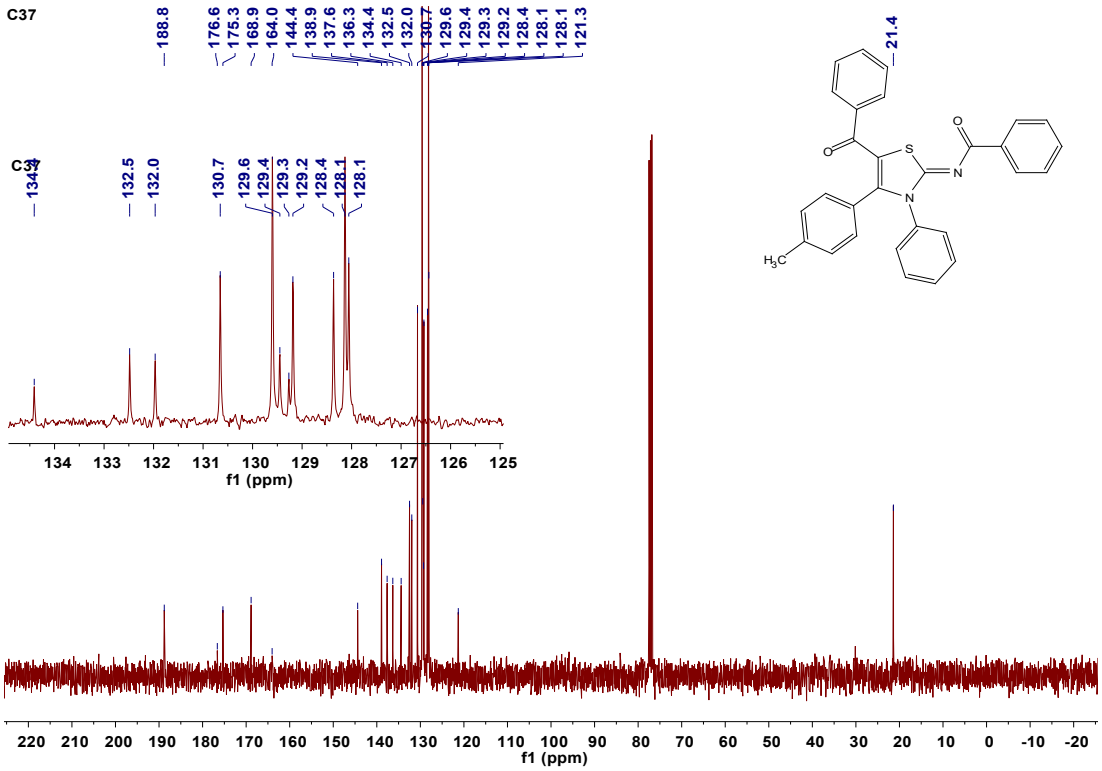


Figure S62 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-phenyl-4-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c37)

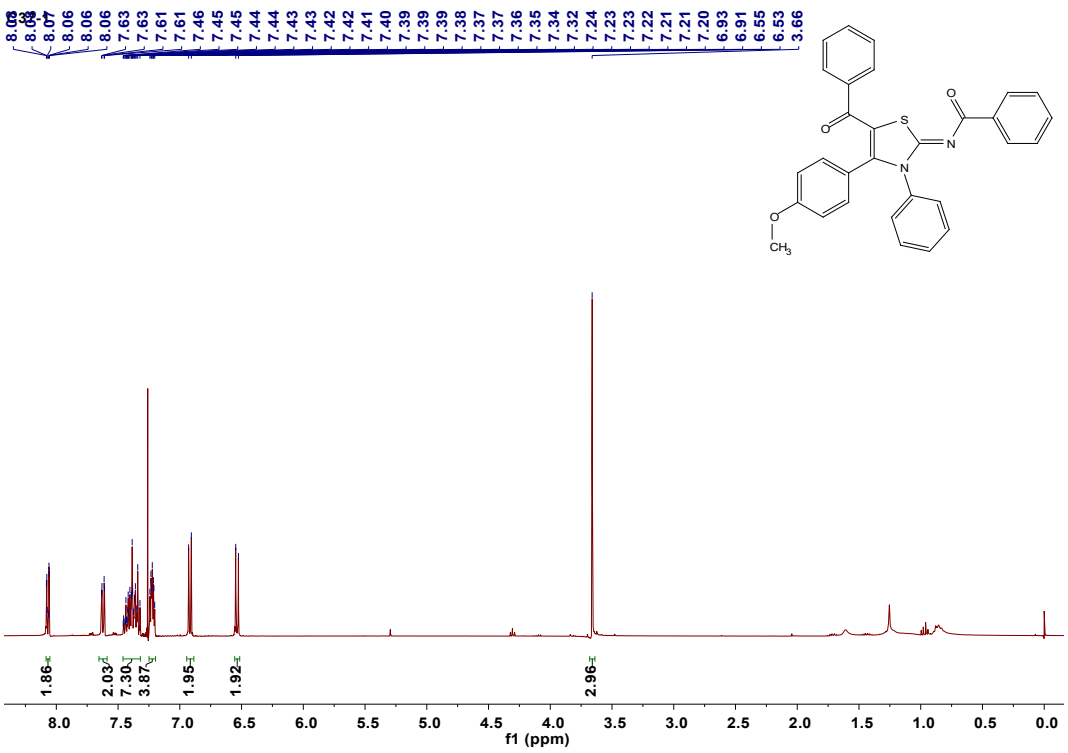


Figure S63 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-phenyl-4-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c37-1)

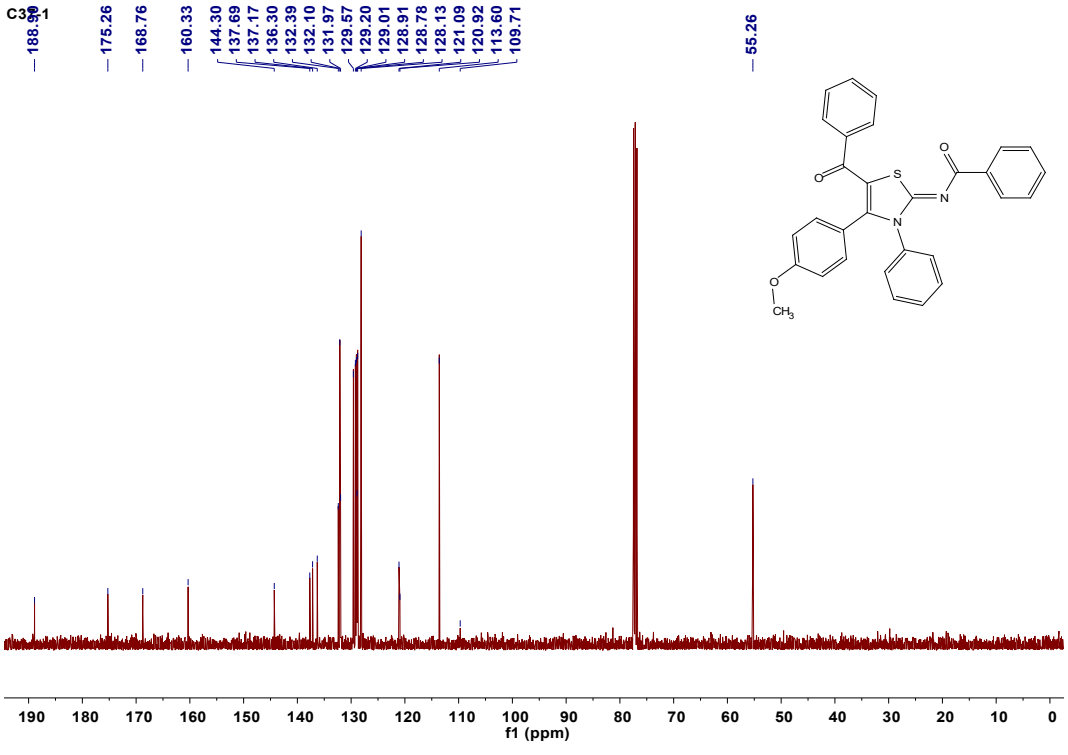


Figure S64 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-benzoyl-3-phenyl-4-(p-tolyl)thiazol-2(3H)-ylidene)benzamide (c37-1)

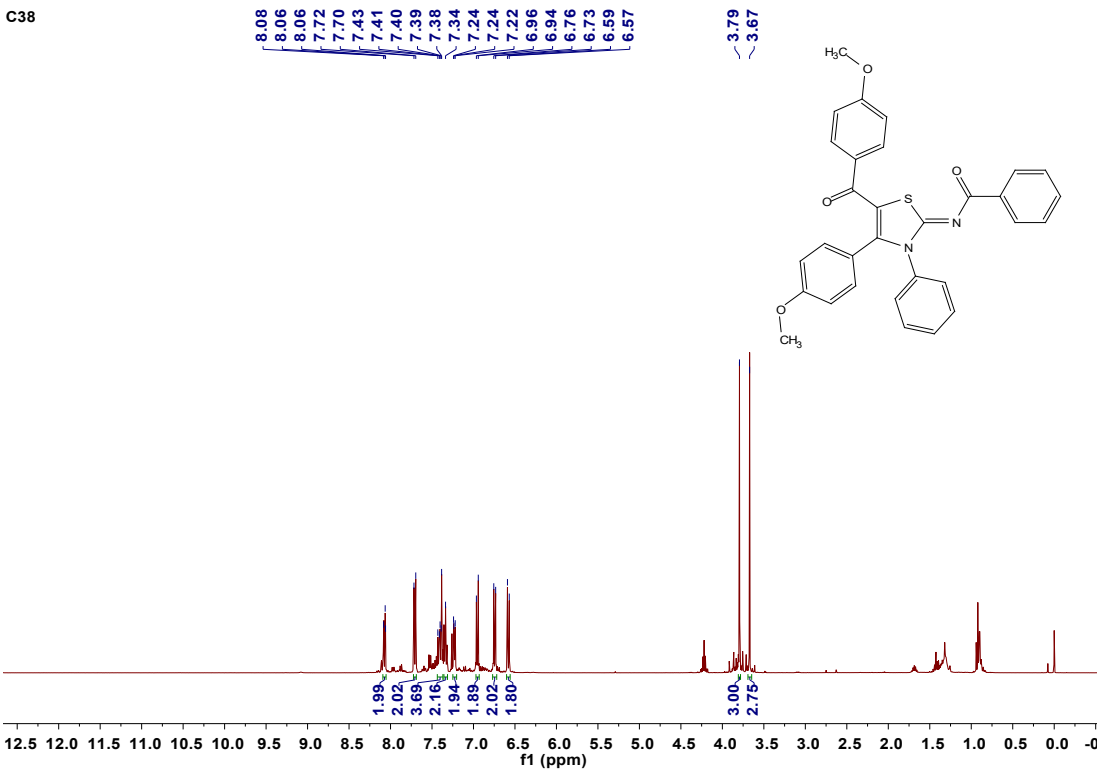


Figure S65 ^{1}H NMR (400 MHz, $CDCl_3$) of *N*-(5-(4-methoxybenzoyl)-4-(4-methoxyphenyl)-3-phenylthiazol-2(3H)-ylidene)benzamide (c38)

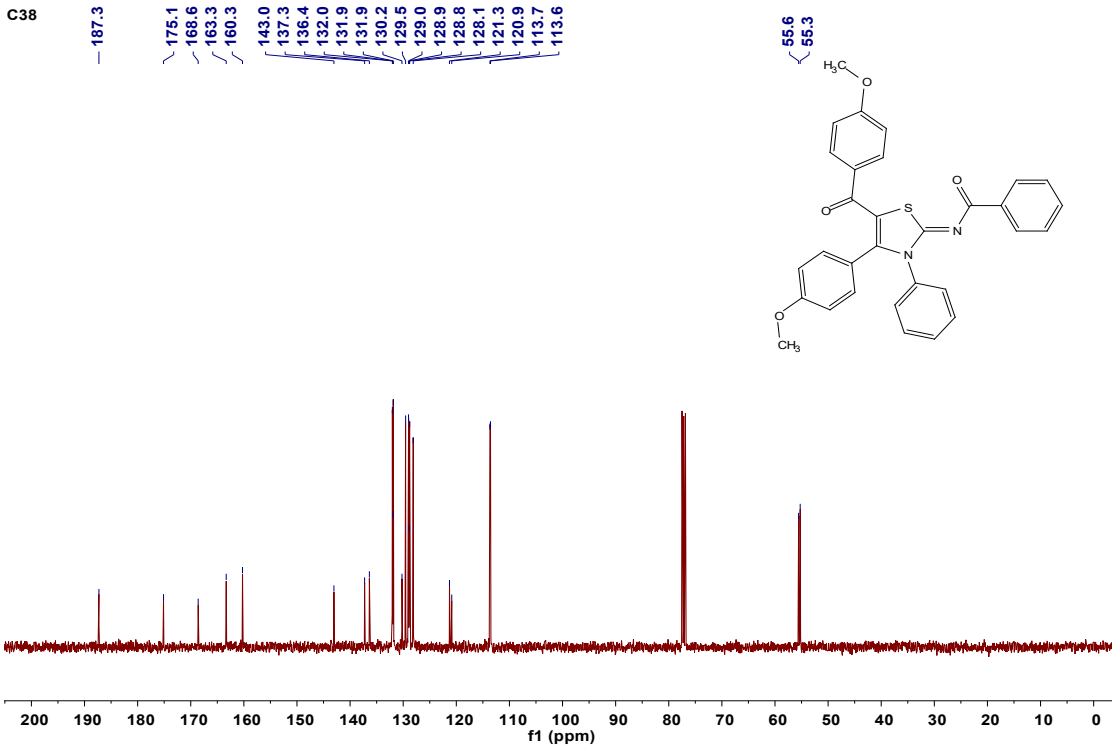


Figure S66 ^{13}C NMR (101 MHz, $CDCl_3$) of *N*-(5-(4-methoxybenzoyl)-4-(4-methoxyphenyl)-3-phenylthiazol-2(3H)-ylidene)benzamide (c38)

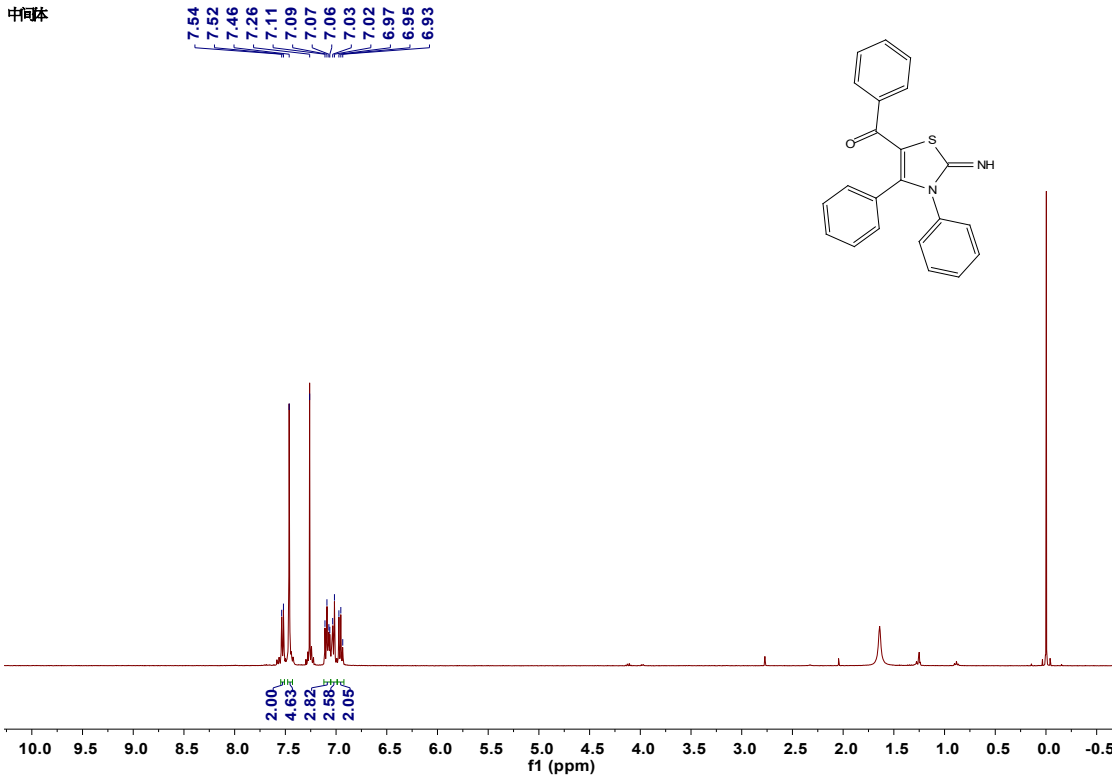


Figure S67 ^{1}H NMR (400 MHz, $CDCl_3$) of (2-imino-3,4-diphenyl-2,3-dihydrothiazol-5-yl)(phenyl)methanone (imine)

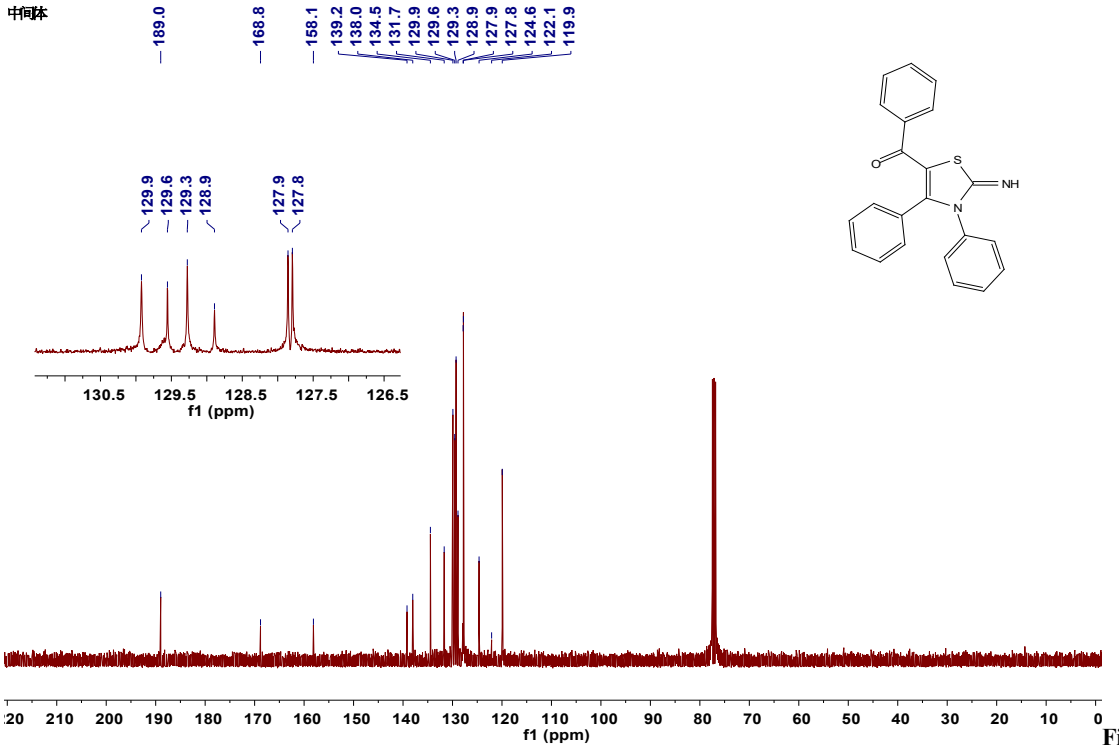


Figure S68 ^{13}C NMR (101 MHz, $CDCl_3$) of (2-imino-3,4-diphenyl-2,3-dihydrothiazol-5-yl)(phenyl)methanone (imine)

6. References

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[2] (a) R. Alfonsi, B. Botta, S. Cacchi, L.-D. Marcotullio, G. Fabrizi, R. Faedda, A. Goggiamani, A. Iazzetti and M. Mori, *J. Med. Chem*., **2017**, *60*, 1469−1477. (b) J. Liu, W. Wei, T. Zhao, X.-Y. Liu, J. Wu, W.-Q. Yu and J.-B. Chang, *J. Org. Chem*., **2016**, *81*, 9326-9336. (c) G.-L. Cheng, X.-B. Zeng, J.-H. Shen, X.-S. Wang and X.-L. Cui, *Angew. Chem., Int. Ed*., **2013**, *52*, 13265−13268. (d) K.-R. Reddy, A.-S. Reddy, R. Shankar, R. kant and P. Das, *Asian J. Org. Chem*., **2015**, *4*, 573−583.

[3] (a) G. M. Sheldrick, SHELXL-97: Program for Crystal Structure Refinement; University of Göttingen: Göttingen, Germany, **1997**. (b) G. M. Sheldrick, SHELXTL Reference Manual, version 5.1; Bruker AXS: Madison, WI, **1997**. (c) G. M. Sheldrick, Acta Crystallogr. **2015**, C71, 3–8.