Supporting Information for

Pd-Catalyzed Multicomponent Reaction of Sulfonyl azide, Primary amines and Methyl α-lsocyanoacetates: Highly Efficient Synthesis of Tetrasubstituted Imidazolone Derivatives

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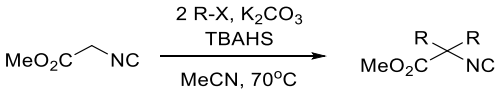
Ⅰ. General Information

Unless otherwise noted, all commercially available compounds were used as provided without further purification. Solvents for chromatography were analytical grade and used without further purification. Anhydrous DMF, was purchased from Beijing InnoChem Science & Technology Co., Ltd. Analytical thin-layer chromatography (TLC) was performed on silica gel, visualized by irradiation with UV light. For column chromatography, 300-400 mesh silica gel was used. ^{1}H-NMR and ^{13}C-NMR were recorded on a BRUKER 400 MHz spectrometer in $CDCl_3$. Chemical shifts (δ) were reported referenced to an internal tetramethylsilane standard or the $CDCl_3$ residual peak (δ 7.26) for ^{1}H NMR. Chemical shifts of ^{13}C NMR are reported relative to $CDCl_3$ (δ 77.16). Data are reported in the following order: chemical shift (δ) in ppm; multiplicities are indicated s (singlet), bs (broad singlet), d (doublet), t (triplet), m (multiplet); coupling constants (J) are in Hertz (Hz). IR spectra were recorded on a BRUKER VERTEX 70 spectrophotometer and are reported in terms of frequency of absorption (cm^{-1}). HRMS spectra were obtained by using BRUKER micrOTOF-Q III instrument with ESI source. The starting materials were isolated by SepaBean machine Flash Chromatography, which purchased from Santai Technologies Inc.

II. Synthesis of Substrates

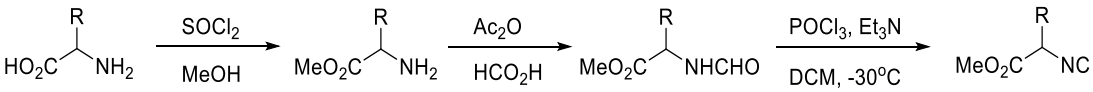
General procedure for the synthesis of Substrate 1. [1]

General procedure A:



To a solution of methyl α-isocyanoacetate (1 equiv) in MeCN (0.1 M) were added K_2CO_3 (4.4 equiv), TBAHS (0.1 equiv) and the corresponding alkyl halide (2 equiv). The reaction mixture was stirred at 70 ℃ for 12 hours. The reaction mixture was cooled to room temperature, quenched with a saturated aqueous solution of $NaHCO_3$ and extracted with DCM (3 times). The combined organic layers were washed with brine, dried over Na_2SO_4, filtered and concentrated under reduced pressure. The crude product was purified by flash chromatography on silica gel to afford the pure product.

General procedure B:

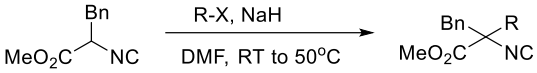


To a solution of Phenylalanine in methanol was added $SOCl_2$. The reaction mixture was stirred at RT for 2 hours and evaporated to dryness to give the corresponding methyl ester hydrochloride which was dissolved in NaOH (3M) and extraceted with DCM (3 times). The combined organic layers were washed with brine, dried over Na_2SO_4, filtered and concentrated under reduced pressure.

The resulting amine was directly used without further purification.
A solution of formic acid (100 equiv) and Ac_2O (10 equiv) was stirred at 60 ℃ for 1 hour. After cooling down the reaction mixture to room temperature the amine was added. The reaction mixture was stirred at RT for 2 hours and evaporated to dryness to give the corresponding formamide which was directly used without further purification.

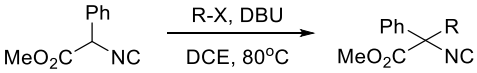
To a solution of the formamide in DCM (0.3 M) was added triethylamine (2.7 equiv). The reaction mixture was cooled to -30 ℃. $POCl_3$ (1.2 equiv) was added and the solution was stirred at -30 ℃ with TLC monitoring (around 3h). The reaction mixture was cooled to room temperature, quenched with a saturated aqueous solution of $NaHCO_3$ and extracted with DCM (3 times). The combined organic layers were washed with brine, dried over Na_2SO_4, filtered and concentrated under reduced pressure. The crude product was purified by flash chromatography on silica gel (Eluent: PE/AcOEt: 9/1 to 7/3) to afford the desired isocyanoacetate.

General procedure C:



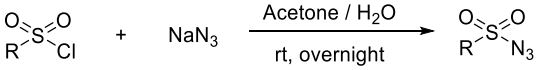
To a solution of methyl 2-isocyano-3-phenylpropanoate (1 equiv) in DMF (0.1 M) was added portionwise NaH (1.5 equiv). After 30 minutes, the corresponding alkyl halide (1.5 equiv) was added dropwise. The reaction mixture was stirred at 50 ℃ for 12 hours. The reaction mixture was cooled to room temperature, quenched with a saturated aqueous solution of $NaHCO_3$ and extracted with DCM (3 times). The combined organic layers were washed with brine, dried over Na_2SO_4, filtered and concentrated under reduced pressure. The crude product was purified by flash chromatography on silica gel to afford the pure product.

General procedure D:



To a solution of methyl 2-isocyano-2-phenylacetate (1 equiv) in DCE (0.1 M) were added DBU (1.1 equiv) and the corresponding alkyl halide (1.5 equiv). The reaction mixture was stirred at 80 ℃ for 12 hours. The reaction mixture was cooled to room temperature, quenched with a saturated aqueous solution of $NaHCO_3$ and extracted with DCM (3 times). The combined organic layers were washed with brine, dried over Na_2SO_4, filtered and concentrated under reduced pressure. The crude product was purified by flash chromatography on silica gel to afford the pure product.

General procedure for the synthesis of sulfonyl azides. [2]



To a solution of sodium azide (2.0 g, 30 mmol) in water (10 mL) was added dropwise over 1h a solution of sulfonyl chloride (20 mmol) in acetone (20 mL) at 0 ℃. The reaction mixture was warmed up to room temperature and stirred for 11 h. Acetone was removed under reduced pressure

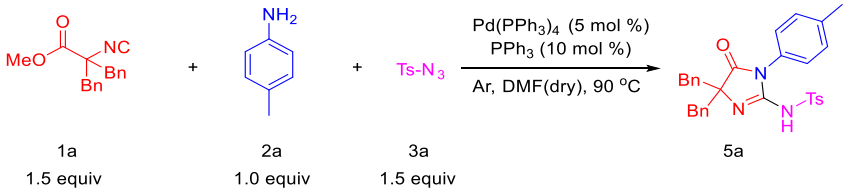
and the reaction mixture was extracted with EtOAc (30 mL x 3). The combined organic layers were dried over $MgSO_4$ and solvent was removed under reduced pressure. Crude product was used without further purification.

III .General Procedure and Product Characterization

1. General Procedure for the Formation of 5a

A representative procedure synthesis of N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (**5a**) is shown below.

In a 25 mL Schlenk tube, to a mixture of methyl 2-benzyl-2-isocyano-3-phenylpropanoate **1a** (0.3 mmol, 1.5 equiv), *p*-toluidine **2a** (0.2 mmol, 1.0 equiv), 4-methylbenzenesulfonyl azide **3a** (0.3 mmol, 1.5 equiv), $Pd(PPh_3)_4$ (5 mol %), PPh_3 (10 mol %) were added in 2 mL anhydrous DMF. The system was under the Ar at 90 ℃ (checked by TLC). After 12h, cooled to rt. The system was extracted with DCM (3 times). The combined organic layers were dried over Na_2SO_4 and solvent was removed under reduced pressure. The residue was purified by flash column chromatography with ethyl acetate and petroleum ether (EA : PE = 1 : 5) as eluents to afford pure product **5a**.



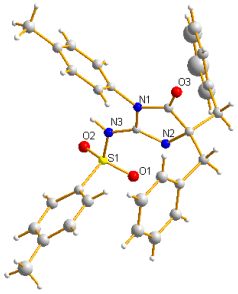
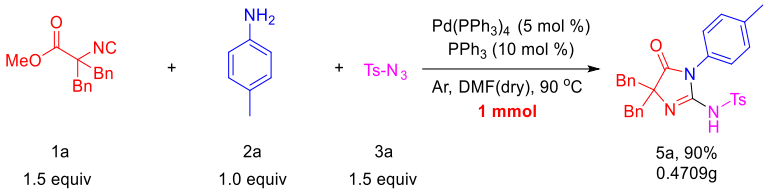


Figure1. Crystal structure of 5a (CCDC 1904592).

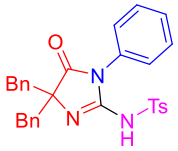
The procedure scale-up synthesis of N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (**5a**) is shown below.

In a 25 mL Schlenk tube, to a mixture of methyl 2-benzyl-2-isocyano-3-phenylpropanoate **1a** (1.5 mmol, 1.5 equiv), *p*-toluidine **2a** (1 mmol, 1.0 equiv), 4-methylbenzenesulfonyl azide **3a** (1.5 mmol, 1.5 equiv), $Pd(PPh_3)_4$ (5 mol %), PPh_3 (10 mol %) were added in 5 mL anhydrous DMF. The system was under the Ar at 90 ℃ (checked by TLC). After 16h, cooled to rt. The system was

extracted with DCM (3 times). The combined organic layers were dried over Na_2SO_4 and solvent was removed under reduced pressure. The residue was purified by flash column chromatography with ethyl acetate and petroleum ether (EA : PE = 1 : 5) as eluents to afford pure product **5a**.

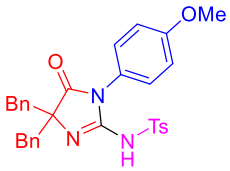


2. Product Characterization



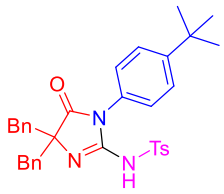
N-(4,4-dibenzyl-5-oxo-1-phenyl-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4a)

Yield: 85% (86.6 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3297, 2988, 2973, 2361, 2340, 1760, 1623, 1394, 1268, 1078, 1052, 894, 685, 668, 604. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.43 (s, 1H), 7.55 (d, *J* = 7.8 Hz, 2H), 7.26 (s, 10H), 7.20 (s, 5H), 6.32 (d, *J* = 7.3 Hz, 2H), 3.33 (d, *J* = 13.6 Hz, 2H), 3.13 (d, *J* = 13.7 Hz, 2H), 2.40 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.3, 155.3, 144.0, 139.5, 133.9, 133.8, 130.4, 130.3, 129.4, 129.0, 128.7, 127.7, 127.2, 126.2, 69.3, 42.8, 21.6. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{28}N_3O_3S(M+H)^+$: 510.1851, found 510.1844.



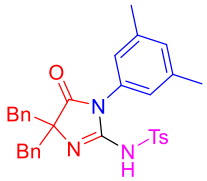
N-(4,4-dibenzyl-1-(4-methoxyphenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4b)

Yield: 94% (101.9 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}):3320, 1763, 1618, 1514, 1451, 1255, 1154, 1119, 1076, 1025, 900, 847, 750, 675. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.49 (s, 1H), 7.56 (d, *J* = 7.8 Hz, 2H), 7.26 (s, 10H), 7.19 (d, *J* = 7.8 Hz, 2H), 6.67 (d, *J* = 8.4 Hz, 2H), 6.20 (d, *J* = 8.4 Hz, 2H), 3.68 (s, 3H), 3.30 (d, *J* = 13.5 Hz, 2H), 3.12 (d, *J* = 13.7 Hz, 2H), 2.39 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.6, 159.7, 155.5, 142.9, 139.5, 133.9, 130.3, 129.4, 128.6, 128.4, 127.7, 126.2, 123.0, 114.3, 69.3, 55.4, 42.7, 21.6. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{31}H_{29}N_3O_4SNa(M+Na)^+$:562.1776, found 562.1750.



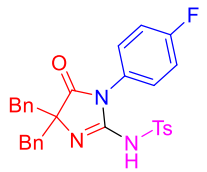
N-(4,4-dibenzyl-1-(4-(tert-butyl)phenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4c)

Yield: 74% (83.5 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3350, 2967, 2360, 2341, 1769, 1623, 1264, 1117, 1071, 900, 852, 753, 662, 602. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.56 (s, 2H), 7.23 (d, *J* = 21.2 Hz, 14H), 6.40–6.17 (m, 2H), 3.33 (d, *J* = 13.7 Hz, 2H), 3.11 (d, *J* = 13.6 Hz, 2H), 2.41 (s, 3H), 1.22 (s, 9H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.5, 155.6, 151.8, 142.9, 139.6, 133.8, 130.3, 129.4, 128.7, 127.7, 126.5, 126.2, 126.0, 100.1, 69.2, 42.8, 34.7, 31.3, 21.7. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{34}H_{36}N_3O_3S$ $(M+H)^+$:566.2477, found 566.2453 .



N-(4,4-dibenzyl-1-(3,5-dimethylphenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4d)

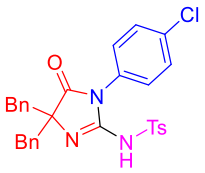
Yield: 57% (59.7 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3336, 2363, 1766, 1616, 1453, 1267,1074, 946, 859, 812, 753, 703, 687, 646, 603. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.35 (s, 1H), 7.56 (d, *J* = 7.9 Hz, 2H), 7.26 (d, *J* = 7.6 Hz, 10H), 7.20 (d, *J* = 7.9 Hz, 2H), 6.84 (s, 1H), 5.85 (s, 2H), 3.31 (d, *J* = 13.6 Hz, 2H), 3.11 (d, *J* = 13.7 Hz, 2H), 2.41 (s, 3H), 2.13 (s, 6H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.5, 155.6, 142.9, 139.6, 138.6, 133.8, 130.9, 130.4, 130.2, 129.3, 128.7, 127.7, 126.2, 124.9, 69.2, 42.8, 21.6, 21.1. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{32}H_{32}N_3O_3S$ $(M+H)^+$:538.2164, found 538.2155 .



N-(4,4-dibenzyl-1-(4-fluorophenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4e)

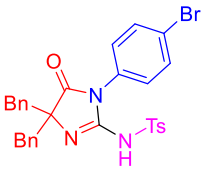
Yield: 79% (83.1 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3314, 2360, 2341, 1759, 1627, 1508, 1392, 1152, 1075, 898, 850, 811, 703, 673. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.46 (s, 1H), 7.56 (d, *J* = 7.9 Hz, 2H), 7.27 (s, 12H), 6.86 (t, *J* = 8.5 Hz, 2H), 6.35–6.20 (m, 2H), 3.33 (d, *J* = 13.6 Hz, 2H), 3.13 (d, *J* = 13.7 Hz, 2H), 2.40 (s,

3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.2, 162.5(*J*=248.2), 155.1, 143.1, 139.4, 133.8, 129.5(0)(*J*=164.8), 129.4(5), 129.1(*J*=8.8), 128.6, 127.8, 126.2, 116.1(*J*=23.0), 69.5, 42.8, 21.7. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{27}FN_3O_3S$ $(M+H)^+$:528.1757, found 528.1748.



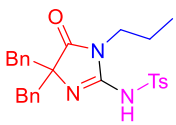
N-(4,4-dibenzyl-1-(4-chlorophenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4f)

Yield: 83% (89.6 mg isolated). White solid. Mp: 207.2 °C -208.1°C. **IR** (neat, ν, cm^{-1}): 3299, 2362, 2341, 1759, 1270, 1119, 1074, 895, 846, 812, 700, 667. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.55 (s, 1H), 7.65 – 7.51 (m, 2H), 7.26 (s, 10H), 7.17 – 7.10 (m, 2H), 6.34 – 6.12 (m, 2H), 3.31 (d, *J* = 13.3 Hz, 2H), 3.13 (d, *J* = 13.8 Hz, 2H), 2.39 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.1, 154.8, 143.2, 139.3, 134.9, 133.8, 130.3, 129.5, 129.2, 128.9, 128.7, 128.5, 127.8, 126.2, 69.6, 42.7, 21.7. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{27}ClN_3O_3S$ $(M+H)^+$:544.1462, found 544.1277.



N-(4,4-dibenzyl-1-(4-bromophenyl)-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4g)

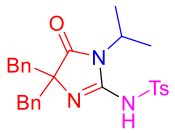
Yield: 67% (78.7 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3295, 2361, 1760, 1625, 1559, 1444, 1386, 1270, 1154, 1116, 1076, 897, 845, 747, 697. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.55 (s, 1H), 7.56 (d, *J* = 8.0 Hz, 2H), 7.25 (s, 14H), 6.17 (d, *J* = 8.4 Hz, 2H), 3.29 (d, *J* = 13.5 Hz, 2H), 3.12 (d, *J* = 13.9 Hz, 2H), 2.38 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.0, 154.7, 143.2, 139.3, 133.8, 132.2, 130.3, 129.5, 129.4, 128.7, 128.6, 127.8, 126.2, 123.0, 69.6, 42.7, 21.7. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{27}BrN_3O_3S$ $(M+H)^+$:588.0957, found 588.0952.



N-(4,4-dibenzyl-5-oxo-1-propyl-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4j)

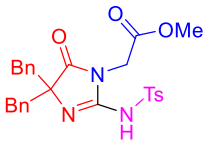
Yield: 48% (62.6 mg isolated). Yellow solid. Mp: 139.6 °C -140.2 °C. **IR** (neat, ν, cm^{-1}):

3313, 2961, 2920, 2359, 1753, 1646, 1593, 1471, 1263, 1145, 1073, 972, 817, 751, 677. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.92 (s, 1H), 7.61 (d, *J* = 7.7 Hz, 2H), 7.23 (d, *J* = 16.5 Hz, 8H), 7.14 (s, 4H), 3.22 (d, *J* = 13.7 Hz, 2H), 3.00 (d, *J* = 12.4 Hz, 4H), 2.43 (s, 3H), 1.60 (s, 2H), 0.35 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 174.0, 155.9, 143.0, 139.5, 133.5, 130.2, 129.4, 128.7, 127.7, 126.3, 68.5, 42.8, 40.8, 21.7, 20.4, 10.8. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{27}H_{29}N_3O_3SNa$ $(M+Na)^+$: 498.1827, found 498.1829.



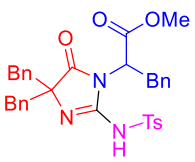
N-(4,4-dibenzyl-1-isopropyl-5-oxo-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (4k)

Yield: 95% (90.3 mg isolated). White solid. Mp: 164.5 ºC -165.5 ºC. **IR** (neat, ν, cm^{-1}): 3334, 1748, 1614, 1495, 1387, 1267, 1066, 944, 869, 750, 702, 676. **^{1}H NMR** (300 MHz, $CDCl_3$) δ 8.22 (s, 1H), 7.61 (d, *J* = 7.9 Hz, 2H), 7.22 (d, *J* = 14.0 Hz, 12H), 3.96 – 3.73 (m, 1H), 3.22 (d, *J* = 13.6 Hz, 2H), 3.02 (d, *J* = 13.7 Hz, 2H), 2.42 (s, 3H), 0.78 (d, *J* = 6.9 Hz, 6H). **^{13}C NMR** (75 MHz, $CDCl_3$) δ 174.1, 155.5, 142.9, 139.8, 133.8, 130.3, 129.4, 128.5, 127.5, 126.2, 68.2, 44.6, 42.7, 21.6, 18.6. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{27}H_{29}N_3O_3SNa$ $(M+Na)^+$:498.1827, found 498.1817.



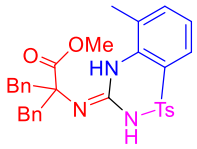
methyl 2-(4,4-dibenzyl-2-((4-methylphenyl)sulfonamido)-5-oxo-4,5-dihydro-1H-imidazol-1-yl)acetate (4l)

Yield: 66% (100.5 mg isolated). White solid. Mp: 176.1 ºC-176.5 ºC. **IR** (neat, ν, cm^{-1}): 3321, 2361, 1760, 1622, 1390, 1270, 1205, 1151, 1119, 1079, 975, 831, 810, 744, 684. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.16 (s, 1H), 7.56 (d, *J* = 7.9 Hz, 2H), 7.29 – 7.12 (m, 12H), 3.79 (s, 2H), 3.32 (s, 3H), 3.24 (d, *J* = 13.7 Hz, 2H), 3.06 (d, *J* = 13.8 Hz, 2H), 2.42 (d, *J* = 13.7 Hz, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.5, 166.3, 154.7, 139.2, 133.5, 130.3, 129.4, 128.7, 127.7, 126.2, 69.1, 52.4, 42.6, 39.8, 21.6. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{27}H_{27}N_3O_5SNa$ $(M+Na)^+$:528.1569, found 528.1562.



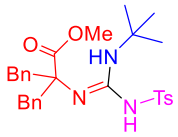
methyl 2-(4,4-dibenzyl-2-((4-methylphenyl)sulfonamido)-5-oxo-4,5-dihydro-1H-imidazol-1-yl)-3-phenylpropanoate (4m)

Yield: 88% (157.0 mg isolated). White solid. Mp: 154.3 °C -155.2 °C. **IR** (neat, ν, cm^{-1}): 3303, 2360, 1760, 1748, 1633, 1494, 1399, 1275, 1228, 1078, 867, 817, 748, 699. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.75 (s, 1H), 7.61 (d, *J* = 7.9 Hz, 2H), 7.37 – 7.29 (m, 3H), 7.26 (d, *J* = 7.3 Hz, 2H), 7.15 (dt, *J* = 18.5, 7.7 Hz, 8H), 7.01 (d, *J* = 7.1 Hz, 4H), 4.75 (t, *J* = 7.8 Hz, 1H), 3.27 (dd, *J* = 14.3, 6.7 Hz, 1H), 3.14 (s, 3H), 3.01 (d, *J* = 13.8 Hz, 1H), 2.82 (dd, *J* = 13.2, 5.1 Hz, 3H), 2.43 (s, 3H), 1.65 (s, 1H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.3, 168.2, 153.9, 143.2, 139.3, 136.7, 133.5, 133.4, 130.5, 129.4, 129.4, 128.9, 128.6, 128.5, 127.9, 127.4, 127.0, 126.4, 67.7, 53.1, 52.4, 42.9, 41.1, 34.5, 21.7. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{34}H_{33}N_3O_5SNa$ $(M+Na)^+$:618.2039, found 618.2052.



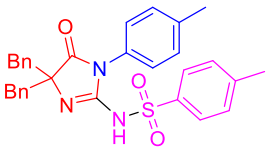
methyl (E)-2-benzyl-2-((5-methyl-1,1-dioxido-1-(p-tolyl)-1,4-dihydro-1l3-benzo[e][1,2,4]thiadiazin-3(2H)-ylidene)amino)-3-phenylpropanoate (4n')

Yield: 90% (103.5 mg isolated). White solid. Mp: 148.7 °C -149.6 °C. **IR** (neat, ν, cm^{-1}): 3326, 2362, 1731, 1547, 1375, 1270, 1217, 1139, 1083, 942, 777, 746, 662. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.76 (s, 1H), 8.00 (d, *J* = 7.9 Hz, 2H), 7.34 (d, *J* = 7.8 Hz, 2H), 7.08 (tt, *J* = 26.9, 7.3 Hz, 7H), 6.89 (d, *J* = 7.6 Hz, 2H), 6.78 (d, *J* = 7.5 Hz, 4H), 5.05 (s, 1H), 3.79 (d, *J* = 13.8 Hz, 2H), 3.66 (s, 3H), 3.28 (d, *J* = 13.8 Hz, 2H), 2.43 (s, 3H), 1.71 (s, 6H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.3, 152.5, 142.4, 141.2, 136.8, 135.6, 131.6, 129.9, 129.5, 128.9, 128.6, 128.5, 127.1, 126.4, 66.9, 52.7, 41.7, 21.6, 17.6. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{33}H_{36}N_3O_4S$ $(M+H)^+$: 570.2427, found 570.2409.



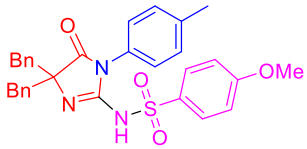
methyl (E)-2-benzyl-2-(((tert-butylamino)((4-methylphenyl)sulfonamido)methylene)amino)-3-phenylpropanoate (4o')

Yield: 95% (99.0 mg isolated). White solid. Mp: 179.0 °C -180.2 °C. **IR** (neat, ν, cm^{-1}): 3419, 3335, 2361, 1734, 1557, 1405, 1140, 1086, 1033, 943, 771, 705, 658. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.92 (d, *J* = 7.9 Hz, 2H), 7.56 (s, 1H), 7.28 (d, *J* = 8.0 Hz, 2H), 7.10 (d, *J* = 6.9 Hz, 6H), 6.80 (d, *J* = 6.9 Hz, 4H), 5.46 (s, 1H), 4.14 (d, *J* = 13.5 Hz, 2H), 3.82 (s, 3H), 3.11 (d, *J* = 13.4 Hz, 2H), 2.38 (s, 3H), 1.01 (s, 9H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.8, 152.3, 142.2, 141.3, 136.0, 129.6, 129.4, 128.2, 127.0, 126.2, 69.0, 52.9, 51.0, 40.9, 29.3, 21.5. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{29}H_{36}N_3O_4S$ $(M+H)^+$:522.2427, found 522.2442.



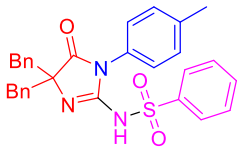
N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (5a)

Yield: 99% (108.2 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3303, 2363, 1760, 1621, 1516, 1436, 1388, 1270, 1154, 1117, 1077, 899, 747, 675. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.40 (s, 1H), 7.55 (d, *J* = 7.7 Hz, 2H), 7.25 (s, 10H), 7.20 (d, *J* = 7.8 Hz, 2H), 6.98 (d, *J* = 7.6 Hz, 2H), 6.20 (d, *J* = 7.7 Hz, 2H), 3.32 (d, *J* = 13.6 Hz, 2H), 3.11 (d, *J* = 13.6 Hz, 2H), 2.40 (s, 3H), 2.23 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.4, 155.5, 142.9, 139.5, 139.0, 133.8, 130.3, 129.7, 129.4, 128.7, 127.8, 127.7, 126.9, 126.2, 69.2, 42.8, 21.6, 21.2. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{31}H_{40}N_3O_3S$ $(M+H)^+$: 524.2008, found 524.2011.



N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methoxybenzenesulfonamide (5b)

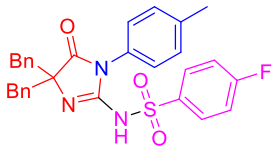
Yield:94% (101.2 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3292, 2987, 2360, 1759, 1634, 1437, 1258, 1120, 1078, 897, 831, 755, 730, 675. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.43 (s, 1H), 7.60 (d, *J* = 8.4 Hz, 2H), 7.26 (s, 10H), 6.97 (d, *J* = 7.8 Hz, 2H), 6.86 (d, *J* = 8.4 Hz, 2H), 6.20 (d, *J* = 7.9 Hz, 2H), 3.82 (s, 3H), 3.31 (d, *J* = 13.6 Hz, 2H), 3.12 (d, *J* = 13.6 Hz, 2H), 2.23 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.5, 162.6, 155.3, 139.0, 134.4, 133.9, 130.3, 129.7, 128.6, 128.3, 127.8, 127.7, 126.9, 113.9, 69.2, 55.7, 42.8, 21.2. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{31}H_{30}N_3O_4S(M+H)^+$:540.1957, found 540.1902.



N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)benzenesulfonamide (5c)

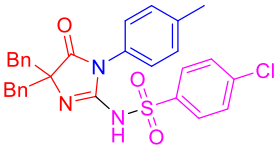
Yield: 60% (61.3 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3312, 2362, 2332, 1761, 1625, 1515, 1390, 1272, 1120, 1076, 898, 846, 717, 652. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.41 (s, 1H), 7.72 (d, *J* = 7.7 Hz, 2H), 7.57 (d, *J* = 8.1 Hz, 1H), 7.47 (t, *J* = 7.9 Hz, 2H), 7.31 (s, 10H), 7.05 (d, *J* = 7.8 Hz, 2H), 6.27 (d, *J* = 8.0 Hz, 2H), 3.40 (d, *J* = 13.5 Hz, 2H), 3.17 (d, *J* = 13.7 Hz, 2H), 2.30 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.4, 155.6, 142.4, 139.1, 133.7, 132.3, 130.3, 129.7, 128.8, 128.7, 127.8,

127.7, 126.9, 126.2, 69.2, 42.8, 21.3. **HRMS** (ESI$^+$, MeCN) m/z calcd for $C_{30}H_{28}N_3O_3S$ $(M+H)^+$:510.1851, found 510.1835.



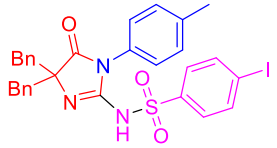
N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-fluorobenzenesulfonamide (5d)

Yield:90% (96.4 mg isolated). White solid. Mp: 196.2°C-197.3°C. **IR** (neat, ν, cm^{-1}): 3322, 2361, 1765, 1625, 1494, 1394, 1239, 1122, 1076, 897, 749, 672. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.36 (s, 1H), 7.64 (dd, *J* = 8.6, 5.0 Hz, 2H), 7.33 – 7.19 (m, 10H), 7.06 (t, *J* = 8.3 Hz, 2H), 6.99 (d, *J* = 7.9 Hz, 2H), 6.20 (d, *J* = 7.9 Hz, 2H), 3.34 (d, *J* = 13.3 Hz, 2H), 3.11 (d, *J* = 13.6 Hz, 2H), 2.24 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.4, 164.8(*J*=252.3), 155.6, 139.2, 138.5(0), 138.4(7), 133.7, 129.7, 129.5(*J*=158.9), 128.8(*J*=9.2), 127.8, 127.6, 126.9, 115.9 (*J*=23.0), 69.3, 42.8, 21.3. **HRMS** (ESI$^+$, MeCN) m/z calcd for $C_{30}H_{27}FN_3O_3S$ $(M+H)^+$:528.1757, found 528.1751.



4-chloro-N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)benzenesulfonamide (5e)

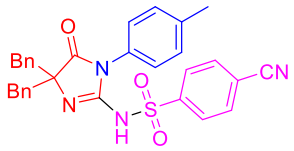
Yield: 83% (90.7 mg isolated). White solid. Mp: 202.3°C-203.1°C. **IR** (neat, ν, cm^{-1}): 3307, 2973, 2362, 1766, 1617, 1515, 1436, 1388, 1269, 1155, 1077, 899, 829, 754, 660. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.34 (s, 1H), 7.56 (d, *J* = 8.1 Hz, 2H), 7.35 (d, *J* = 8.2 Hz, 2H), 7.25 (q, *J* = 7.5, 7.1 Hz, 10H), 6.99 (d, *J* = 7.9 Hz, 2H), 6.19 (d, *J* = 7.9 Hz, 2H), 3.34 (d, *J* = 13.7 Hz, 2H), 3.10 (d, *J* = 13.7 Hz, 2H), 2.24 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.3, 155.7, 140.8, 139.2, 138.6, 133.7, 130.3, 129.8, 129.0, 128.7, 127.8, 127.7, 127.6, 126.9, 69.4, 42.8, 21.3. **HRMS** (ESI$^+$, MeCN) m/z calcd for $C_{30}H_{26}ClN_3O_3SNa$ $(M+Na)^+$:566.1281, found 566.1271.



N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-iodobenzenesulfonamide (5f)

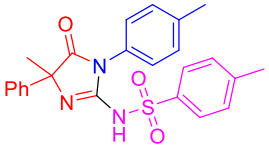
Yield: 71% (89.9 mg isolated). White solid. Mp: >210°C. **IR** (neat, ν, cm^{-1}): 3350, 2987, 2361, 1760, 1619, 1438, 1386, 1263, 1073, 1007, 897, 725, 601. **^{1}H NMR** (400 MHz,

$CDCl_3$) δ 8.25 (s, 1H), 7.78 – 7.71 (m, 2H), 7.36 – 7.32 (m, 2H), 7.30 – 7.27 (m, 3H), 7.25 – 7.24 (m, 1H), 7.22 (dd, *J* = 7.7, 1.9 Hz, 4H), 7.00 (d, *J* = 8.0 Hz, 2H), 6.22 – 6.17 (m, 2H), 3.35 (d, *J* = 13.7 Hz, 2H), 3.10 (d, *J* = 13.7 Hz, 2H), 2.25 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.3, 155.7, 142.0, 139.3, 138.0, 133.6, 130.3, 129.8, 128.7, 127.9, 127.7, 127.6, 126.9, 99.5, 69.3, 42.9, 21.3. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{27}IN_3O_3S$ $(M+H)^+$:636.0818, found 636.0828.



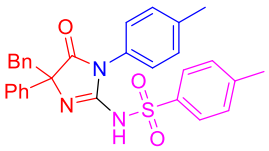
4-cyano-N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)benzenesulfonamide (5g)

Yield: 71% (76.2 mg isolated). White solid. Mp: 205.6 ºC -206.2ºC. **IR** (neat, ν, cm^{-1}): 3340, 2362, 1760, 1626, 1515, 1451, 1388, 1278, 1152, 1076, 900, 848, 754, 646. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.26 (s, 1H), 7.68 (s, 4H), 7.27 (tt, *J* = 13.5, 7.0 Hz, 10H), 7.00 (d, *J* = 7.9 Hz, 2H), 6.19 (d, *J* = 7.9 Hz, 2H), 3.38 (d, *J* = 13.7 Hz, 2H), 3.11 (d, *J* = 13.7 Hz, 2H), 2.25 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 219.2, 173.2, 156.0, 146.3, 139.4, 133.6, 132.6, 130.2, 129.8, 128.8, 127.9, 127.4, 126.8, 117.6, 115.9, 107.8, 69.4, 42.8, 21.3. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{31}H_{27}N_4O_3S$ $(M+H)^+$:535.1804 found 535.1770.



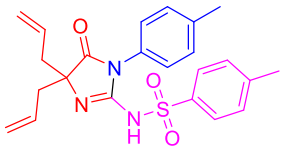
4-methyl-N-(4-methyl-5-oxo-4-phenyl-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)benzenesulfonamide (6d)

Yield: 68% (59.2 mg isolated). White solid. Mp: >210ºC. **IR** (neat, ν, cm^{-1}): 3313, 2360, 1760, 1628, 1515, 1440, 1379, 1264, 1125, 1076, 915, 879, 785, 740, 676. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.30 (s, 1H), 7.71 (d, *J* = 7.8 Hz, 2H), 7.42 (d, *J* = 7.1 Hz, 2H), 7.32 (d, *J* = 7.7 Hz, 3H), 7.20 (d, *J* = 8.4 Hz, 2H), 7.11 (d, *J* = 7.8 Hz, 2H), 7.01 (d, *J* = 8.1 Hz, 2H), 2.35 (s, 3H), 2.27 (s, 3H), 1.87 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.4, 155.3, 143.3, 139.2, 139.1, 137.7, 129.8, 129.6, 129.3, 129.0, 128.2, 127.0, 126.4, 125.2, 64.7, 25.6, 21.7, 21.3. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{24}H_{23}N_3O_3SNa$ $(M+Na)^+$:456.1358 found 456.1332.



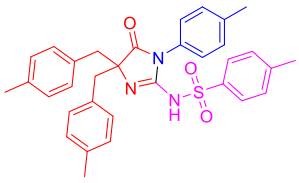
N-(4-benzyl-5-oxo-4-phenyl-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (6e)

Yield: 75% (76.8 mg isolated). White solid. Mp: 209.2 ºC -209.8ºC. **IR** (neat, ν, cm^{-1}): 3303, 2362, 1758, 1635, 1433, 1264, 1138, 1112, 1077, 891, 692, 670, 644. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.57 (s, 1H), 7.65 (dd, *J* = 13.1, 7.6 Hz, 4H), 7.42 (q, *J* = 8.0, 7.6 Hz, 3H), 7.24 (h, *J* = 9.4, 7.8 Hz, 7H), 7.06 (d, *J* = 8.0 Hz, 2H), 6.56 (d, *J* = 7.9 Hz, 2H), 3.68 (d, *J* = 13.6 Hz, 1H), 3.30 (d, *J* = 13.6 Hz, 1H), 2.42 (s, 3H), 2.28 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 172.4, 155.4, 144.2, 143.1, 139.3, 139.1, 136.7, 133.4, 130.6, 129.7, 129.4, 129.2, 129.1, 128.7, 127.9, 127.1, 126.4, 125.7, 69.1, 45.4, 21.7, 21.3. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{30}H_{27}N_3O_3SNa$ $(M+Na)^+$:532.1671 found532.1676.



N-(4,4-diallyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide (6f)

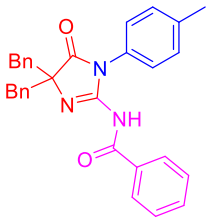
Yield: 48% (40.5 mg isolated). White solid. Mp: 181.7 ºC -182.6ºC. **IR** (neat, ν, cm^{-1}): 3291, 2982, 2360, 1763, 1608, 1380, 1279, 1122, 1079, 891, 844, 814, 731, 677. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.02 (s, 1H), 7.76 (d, *J* = 7.9 Hz, 2H), 7.29 – 7.23 (m, 2H), 7.18 (d, *J* = 7.9 Hz, 2H), 7.00 (d, *J* = 7.9 Hz, 2H), 5.67 (dd, *J* = 16.9, 9.0 Hz, 2H), 5.21 (t, *J* = 12.0 Hz, 4H), 2.58 (qd, *J* = 13.9, 7.4 Hz, 4H), 2.38 (d, *J* = 25.8 Hz, 6H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 173.7, 156.0, 143.2, 139.4, 139.2, 129.8, 129.6, 129.5, 128.2, 127.1, 126.4, 121.9, 66.5, 40.8, 21.7, 21.4. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{23}H_{25}N_3O_3SNa$ $(M+Na)^+$:446.1514 found 446.1531.



N-(4,4-bis(4-methylbenzyl)-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)-4-methylbenzenesulfonamide(6g)

Yield: 69% (113.7 mg isolated). White solid. Mp: >210ºC. **IR** (neat, ν, cm^{-1}): 3305, 1759, 1637, 1514, 1441, 1269, 1153, 1077, 899, 736, 672. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 8.17 (s, 1H), 7.56 (d, *J* = 7.9 Hz, 2H), 7.21 (d, *J* = 7.9 Hz, 2H), 7.12 – 6.96 (m, 10H), 6.27 (d, *J* = 7.8 Hz, 2H), 3.27 (d, *J* = 13.7 Hz, 2H), 3.03 (d, *J* = 13.7 Hz, 2H), 2.42 (s,

3H), 2.28 (d, *J* = 16.3 Hz, 9H). **^{13}C NMR** (100 MHz, $CDCl_3$) δ 173.6, 155.5, 142.8, 139.7, 139.0, 137.3, 130.6, 130.1, 129.6, 129.4, 129.3, 127.9, 127.0, 126.3, 69.1, 42.4, 21.7, 21.3, 21.2. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{33}H_{32}N_3O_3SNa$ $(M+Na)^+$:574.2140 found 574.2150.



N-(4,4-dibenzyl-5-oxo-1-(p-tolyl)-4,5-dihydro-1H-imidazol-2-yl)benzamide (7a)
Yield: 59% (56.2 mg isolated). White solid. Mp: 138.6-139.2. **IR** (neat, ν, cm^{-1}): 3291, 1743, 1631, 1584, 1513, 1443, 1323, 1260, 1168, 1098, 792, 700, 602. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 9.84 (s, 1H), 7.97 – 7.90 (m, 2H), 7.45 – 7.39 (m, 1H), 7.31 (d, *J* = 7.8 Hz, 2H), 7.28 (d, *J* = 1.9 Hz, 2H), 7.26 (d, *J* = 1.6 Hz, 6H), 7.25 (d, *J* = 2.2 Hz, 1H), 7.17 (tq, *J* = 4.1, 2.5, 1.9 Hz, 1H), 7.12 (d, *J* = 8.0 Hz, 2H), 6.54 – 6.48 (m, 2H), 3.40 (d, *J* = 13.6 Hz, 2H), 3.11 (d, *J* = 13.7 Hz, 2H), 2.34 (s, 3H). **^{13}C NMR** (100 MHz, $CDCl_3$)δ 179.3, 174.1, 160.2, 138.6, 136.8, 133.8, 132.2, 130.3, 130.0, 129.7, 129.5, 128.7, 128.1, 127.8, 127.0, 68.8, 43.1, 21.4. **HRMS** (ESI^+, MeCN) m/z calcd for $C_{31}H_{27}N_3O_2Na$ $(M+Na)^+$:496.2001 found 496.2018.

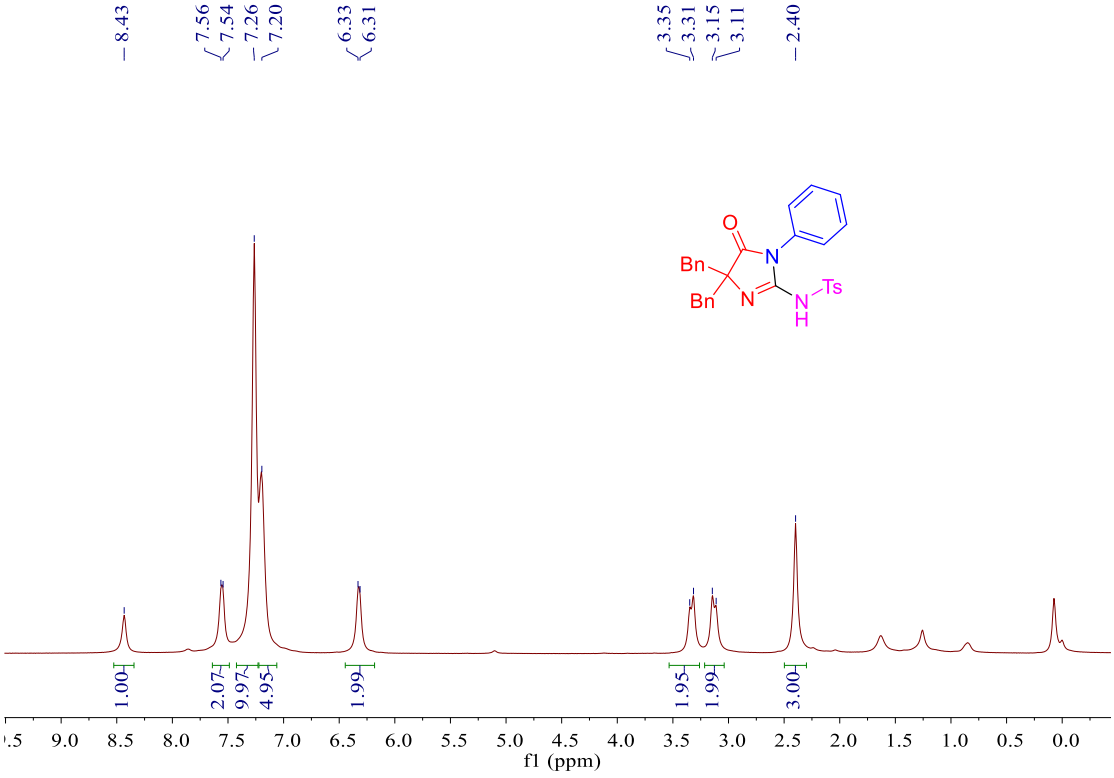
IV. References

(1) (a) [1] A. Clemenceau, Q.Wang, J. Zhu, *Org. Lett.* **2017**, *19*, 4872 – 4875.
(b) A. Clemenceau, Q. Wang, J. Zhu, *Org. Lett*. **2018**, *20*, 126−129
(c) Ito, Y.; Higuchi, N.; Murakami, M. *Tetrahedron Lett*. **1988**, *29*, 5151.
(d) N. Elders, R. F. Schmitz, F. J. J. de Kanter, E. Ruijter, M. B. Groen, R. V. A. Orru. *J. Org. Chem.* **2007**, *72*, 6135-6142.

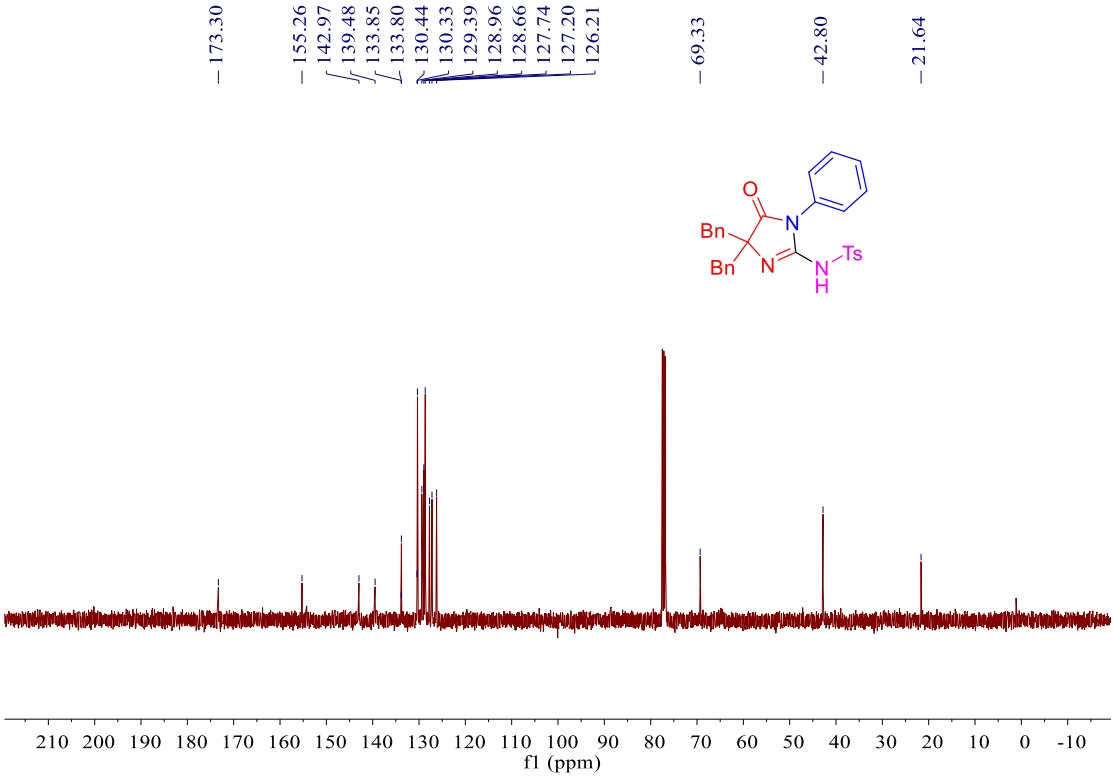
(2) (a) Z.-Y, Gu, Y. Liu, F. Wang, X.-G, Bao, S.- Y, Wang, S. -J Ji, *ACS Catal*. **2017**, *7*, 3893−3899
(b) T. Jiang, Z. -Y, Gu, L. Yin, S. –Y, Wang, S. –J, Ji, *J. Org. Chem.* **2017**, *82*, 7913−7919.

V. Copies of ^{1}H NMR and ^{13}C NMR Spectra

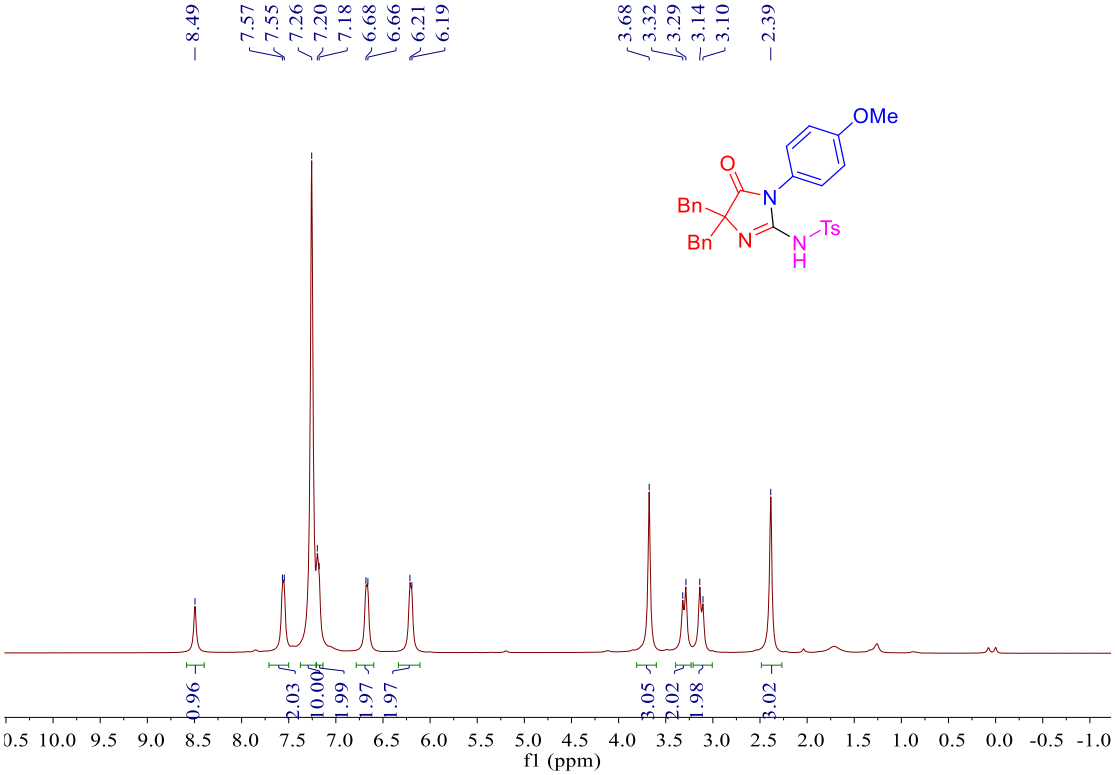
^{1}H NMR Spectra of **4a**



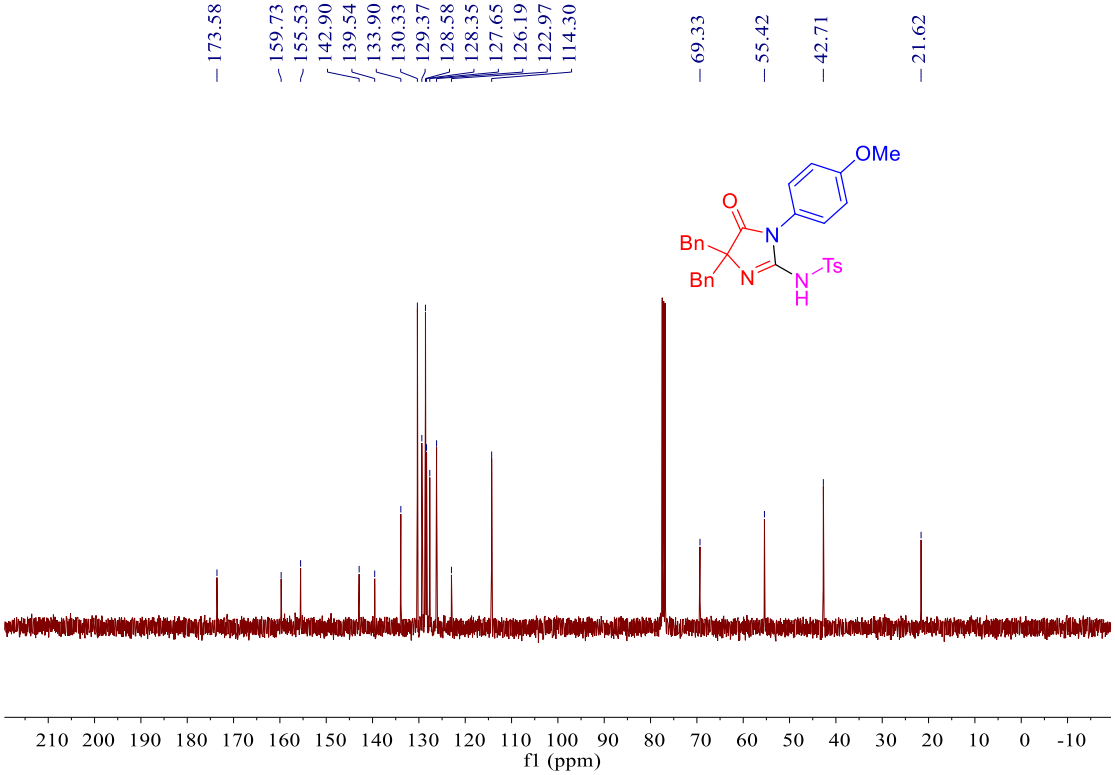
^{13}C NMR Spectra of **4a**



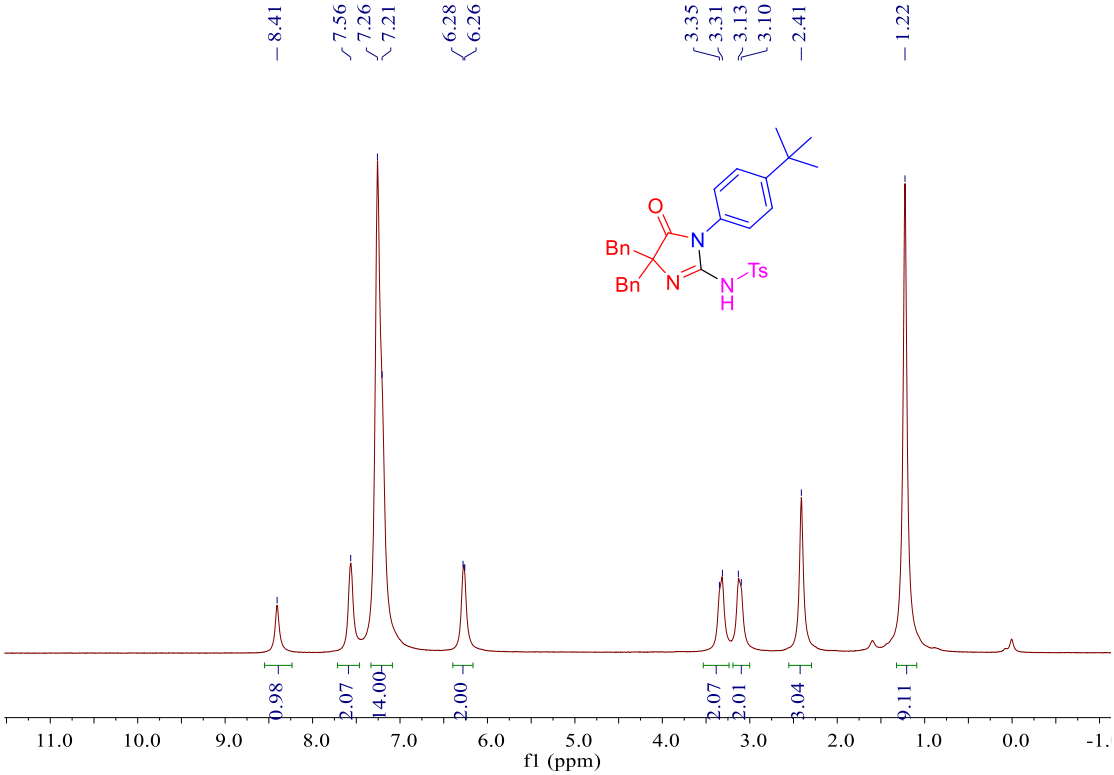
^{1}H NMR Spectra of **4b**



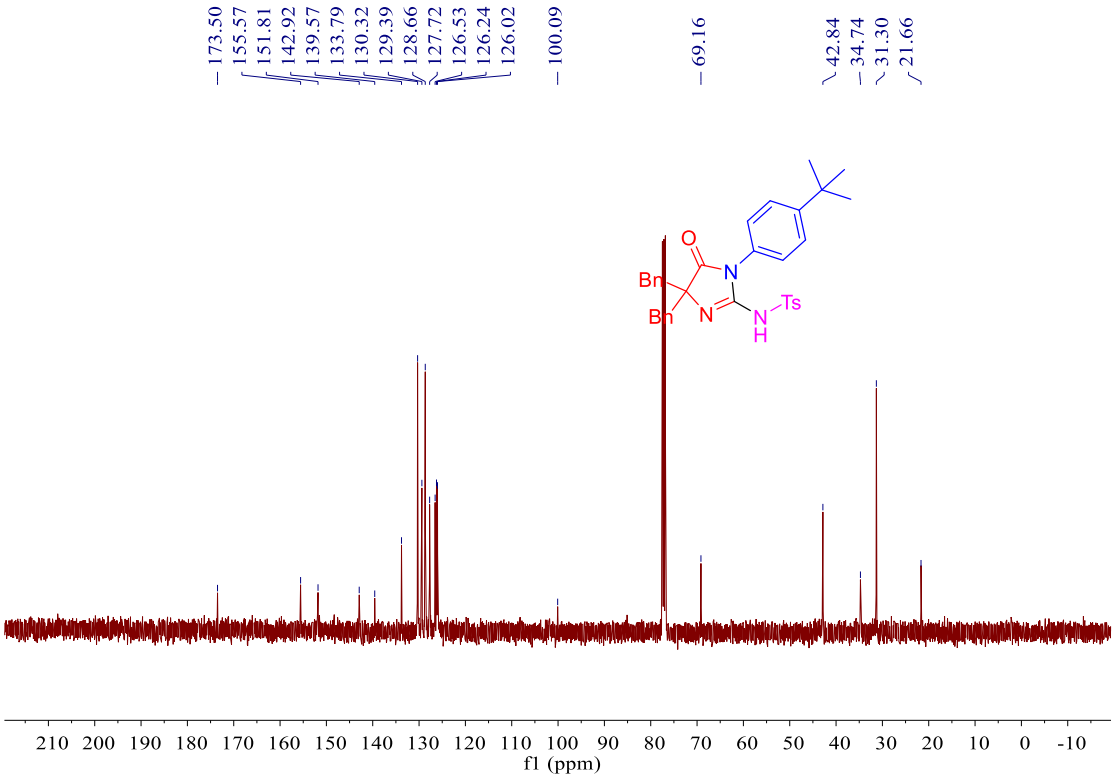
^{13}C NMR Spectra of **4b**



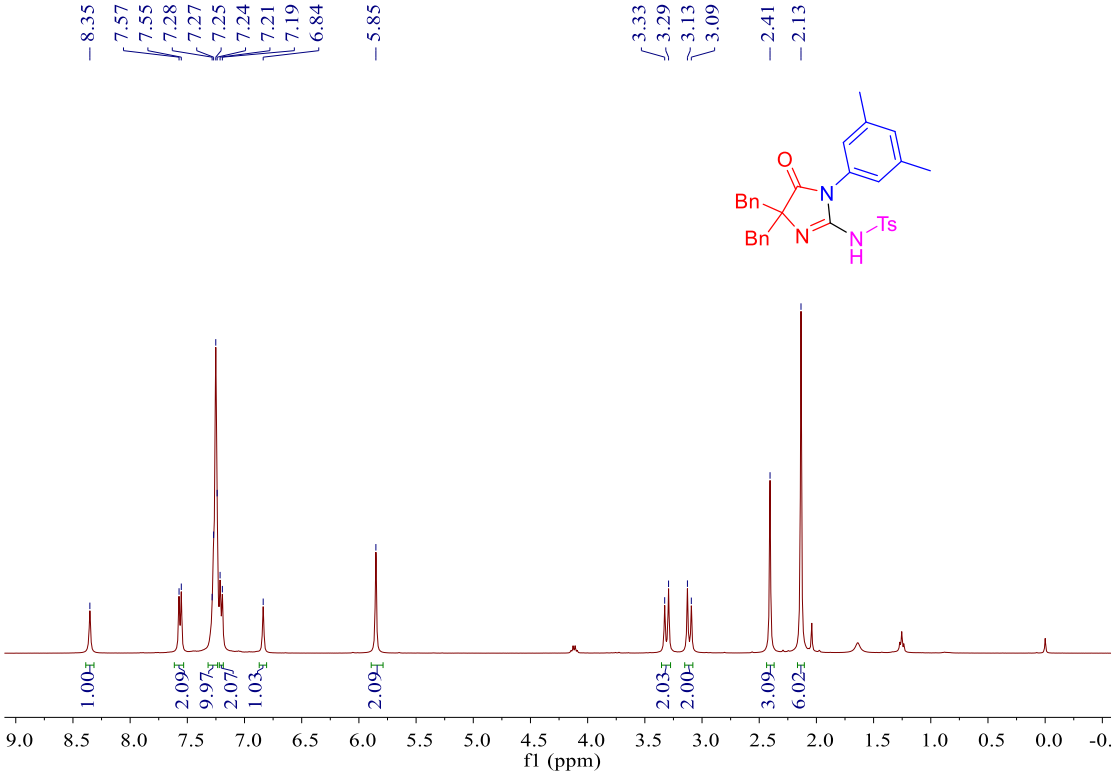
^{1}H NMR Spectra of **4c**



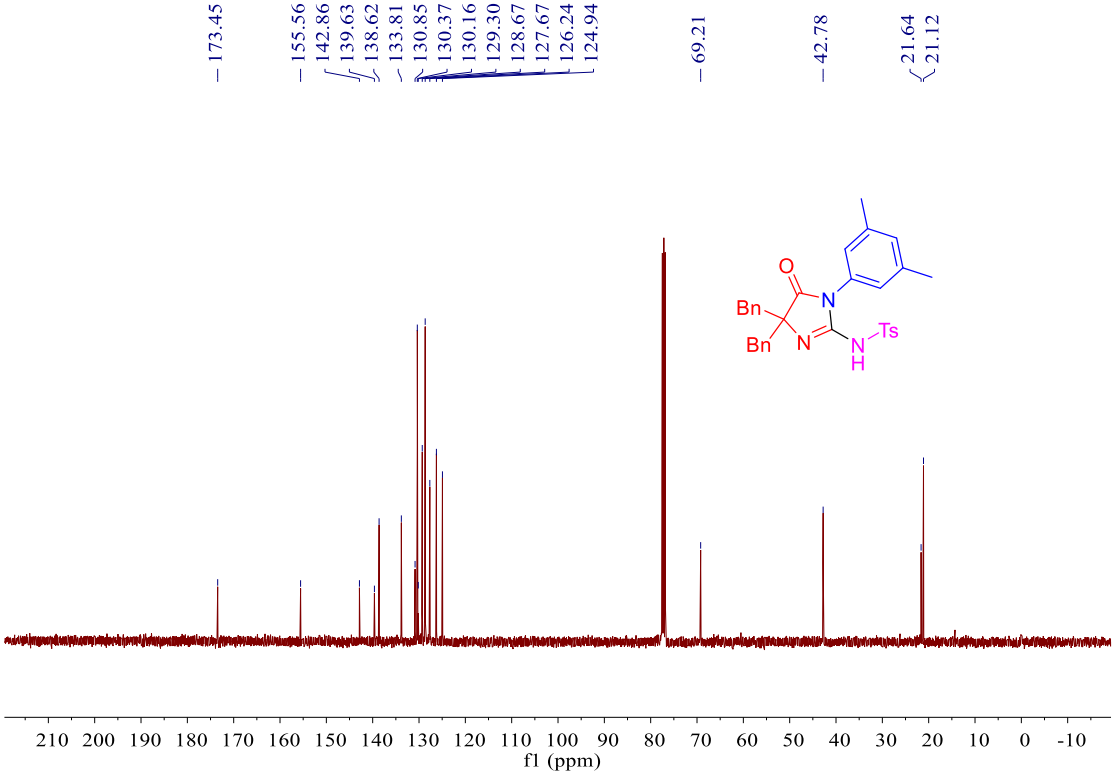
^{13}C NMR Spectra of **4c**



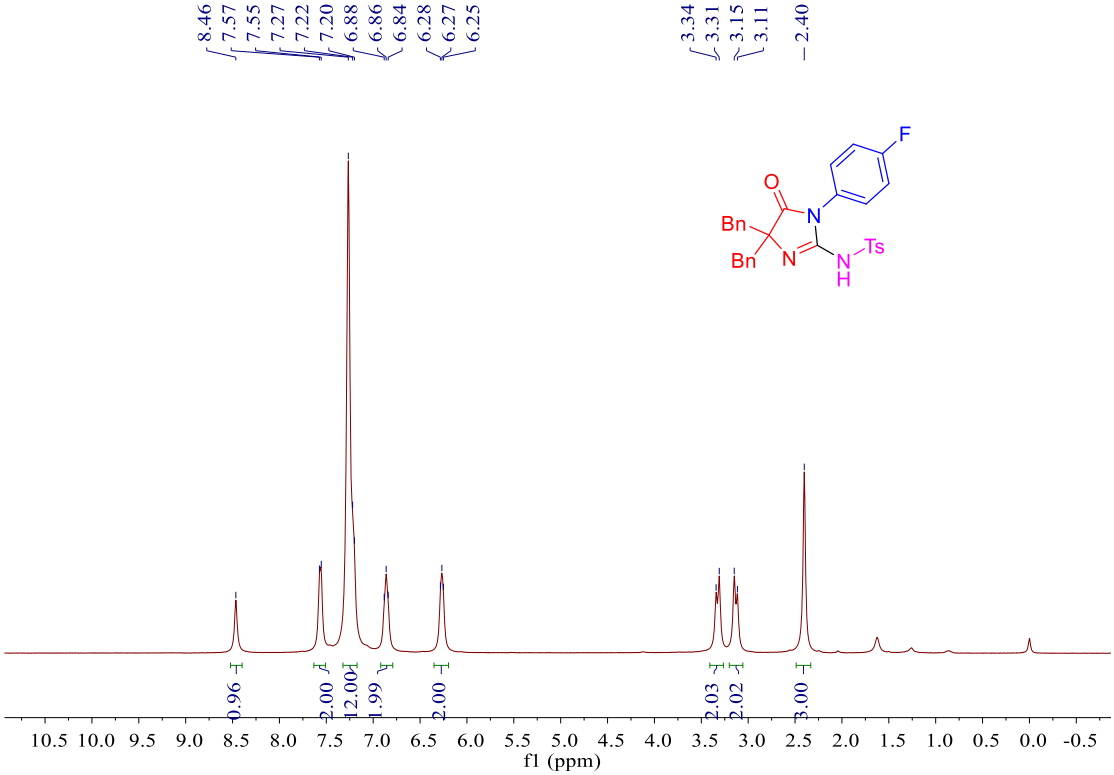
^{1}H NMR Spectra of **4d**



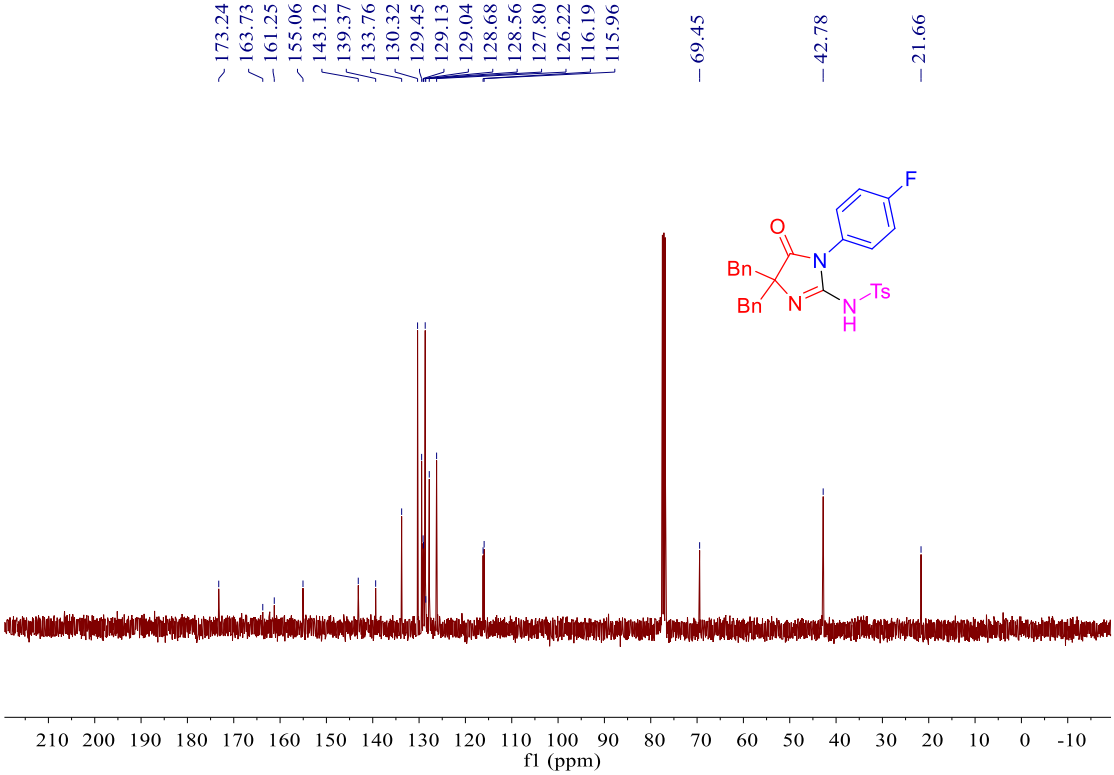
^{13}C NMR Spectra of **4d**



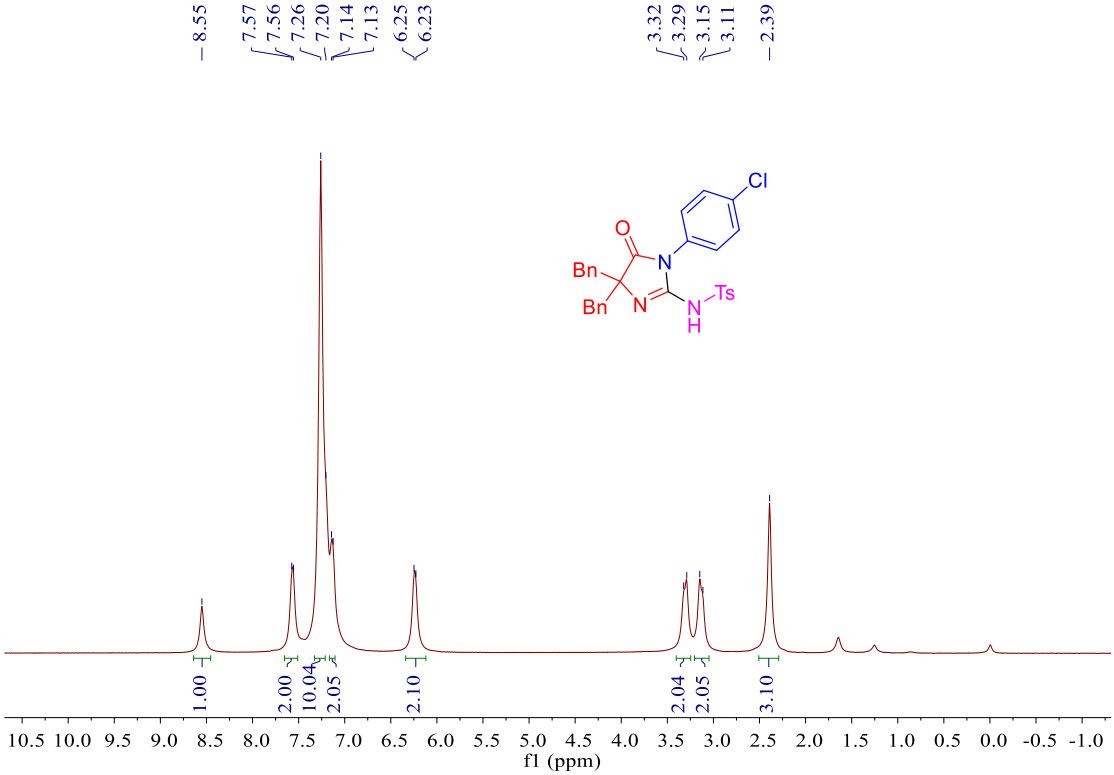
^{1}H NMR Spectra of **4e**



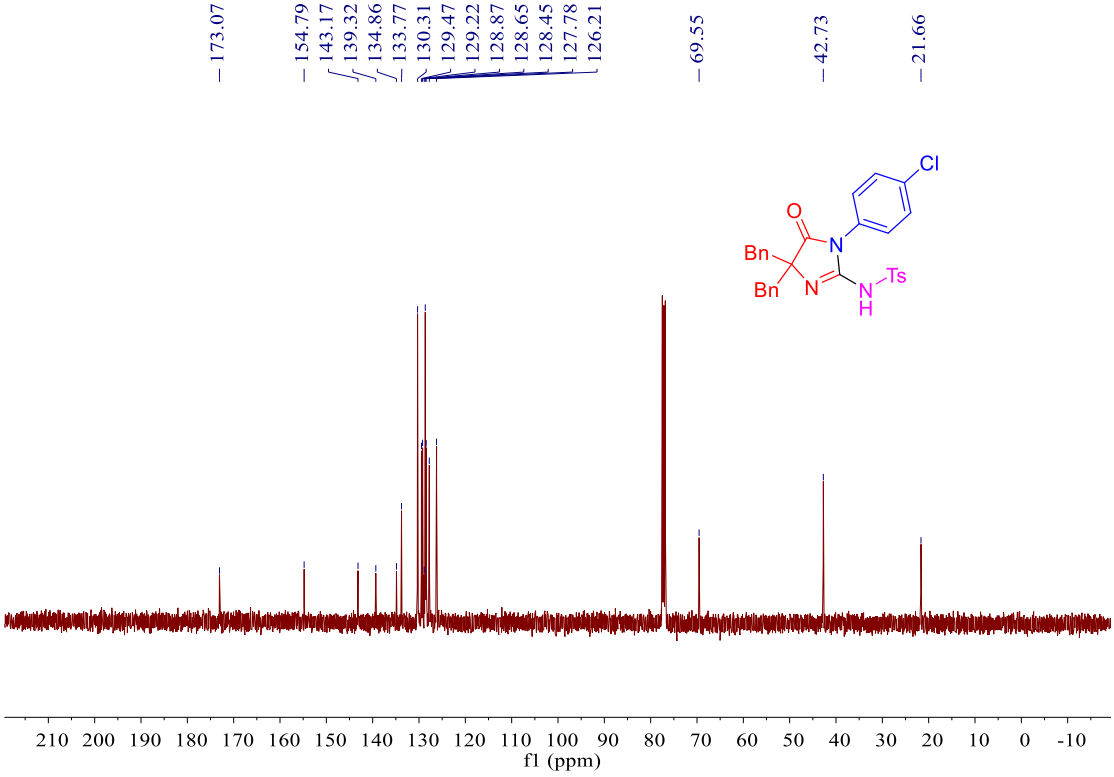
^{13}C NMR Spectra of **4e**



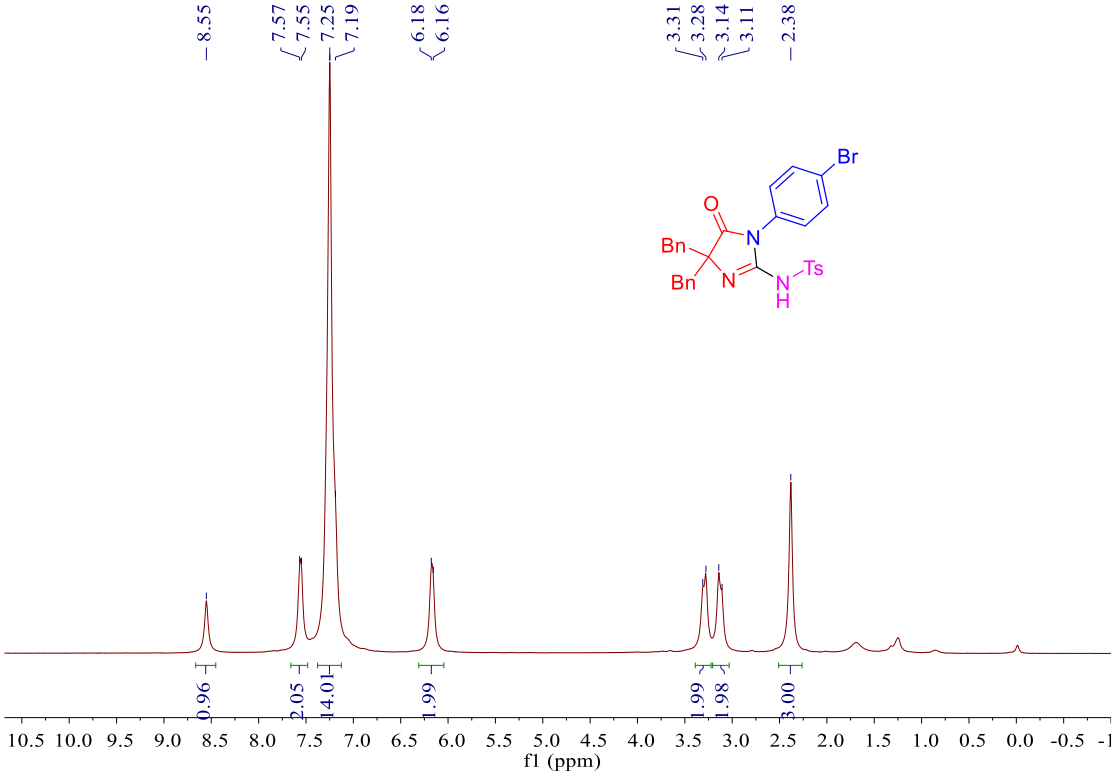
^{1}H NMR Spectra of **4f**



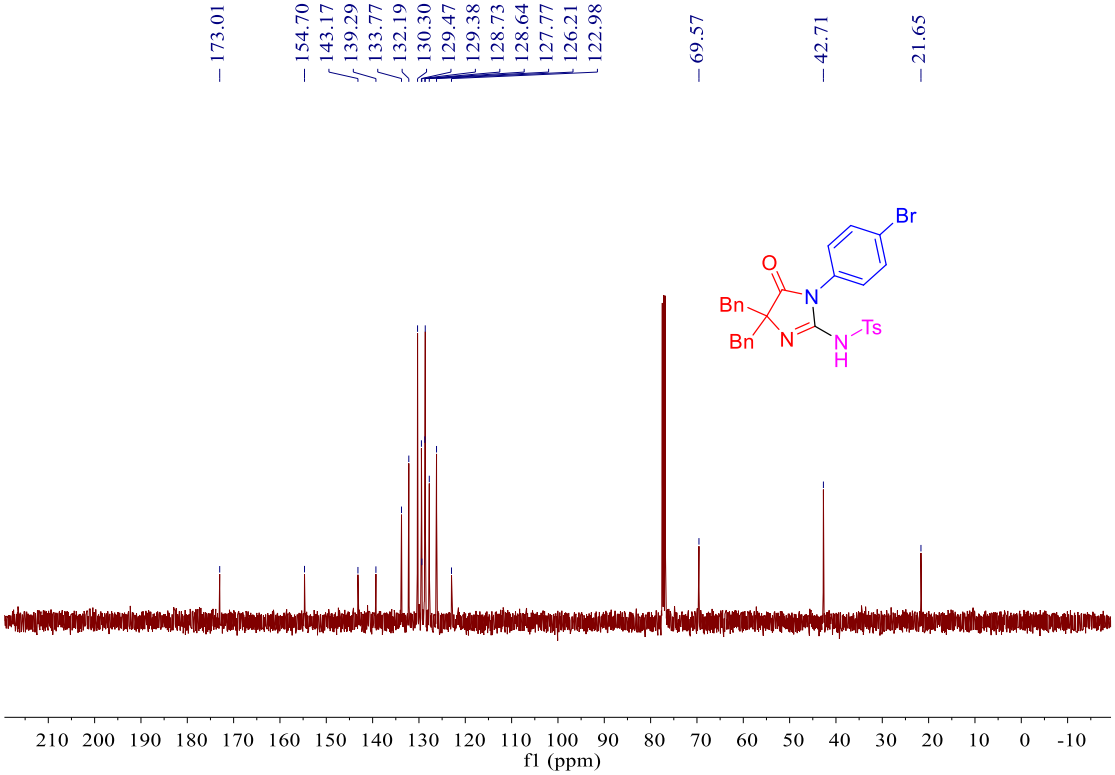
^{13}C NMR Spectra of **4f**



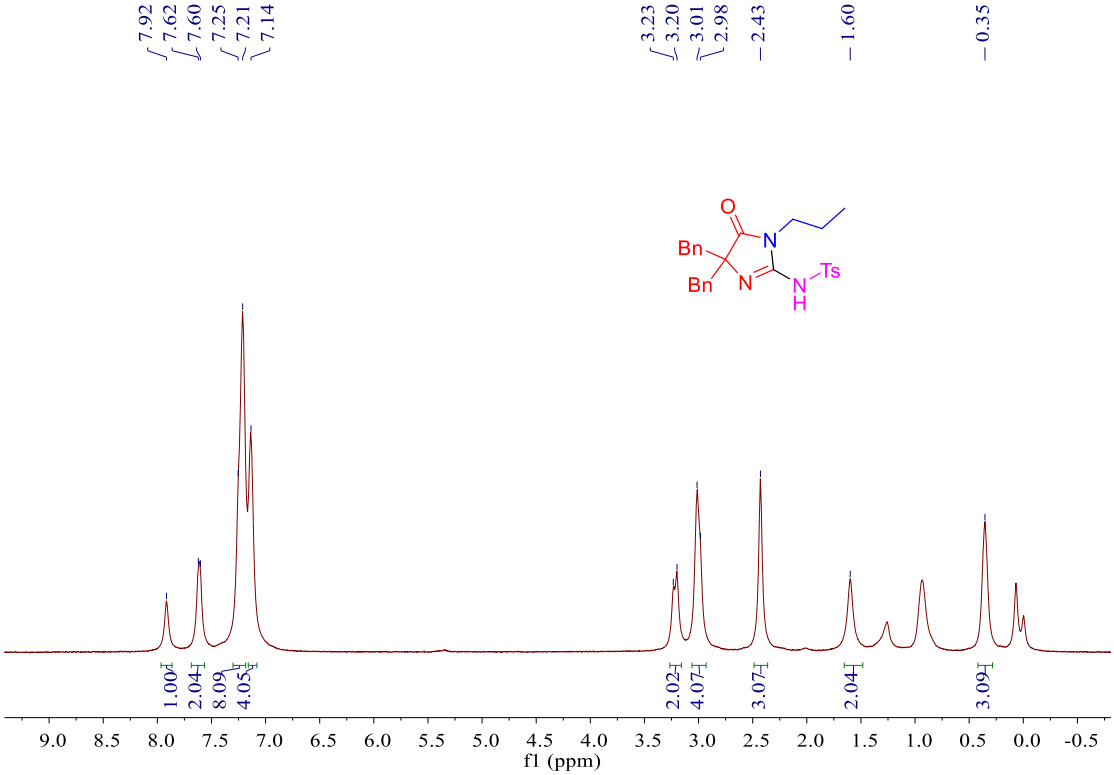
^{1}H NMR Spectra of **4g**



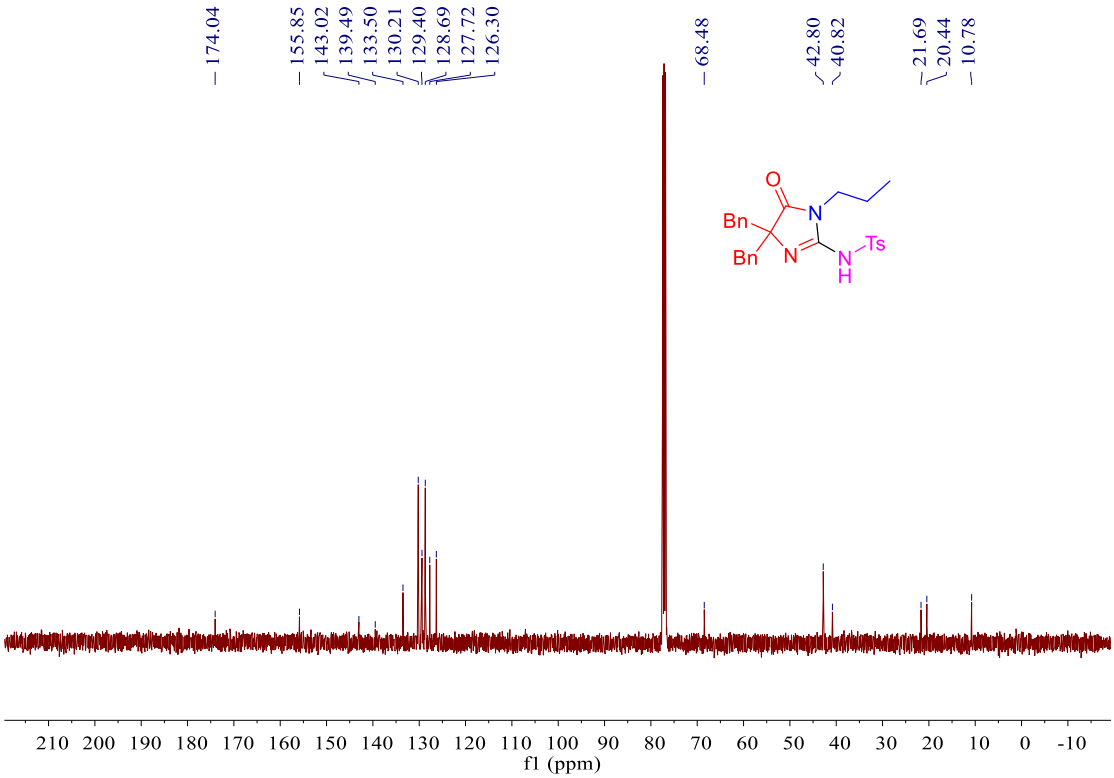
^{13}C NMR Spectra of **4g**



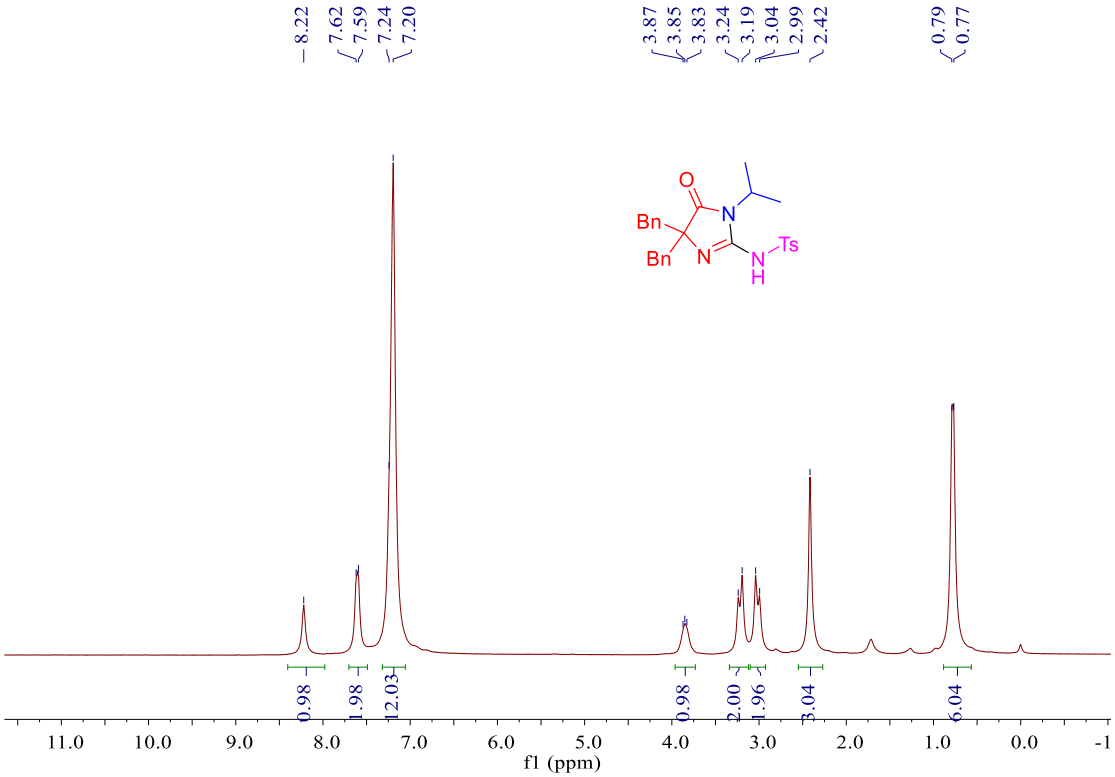
^{1}H NMR Spectra of **4j**



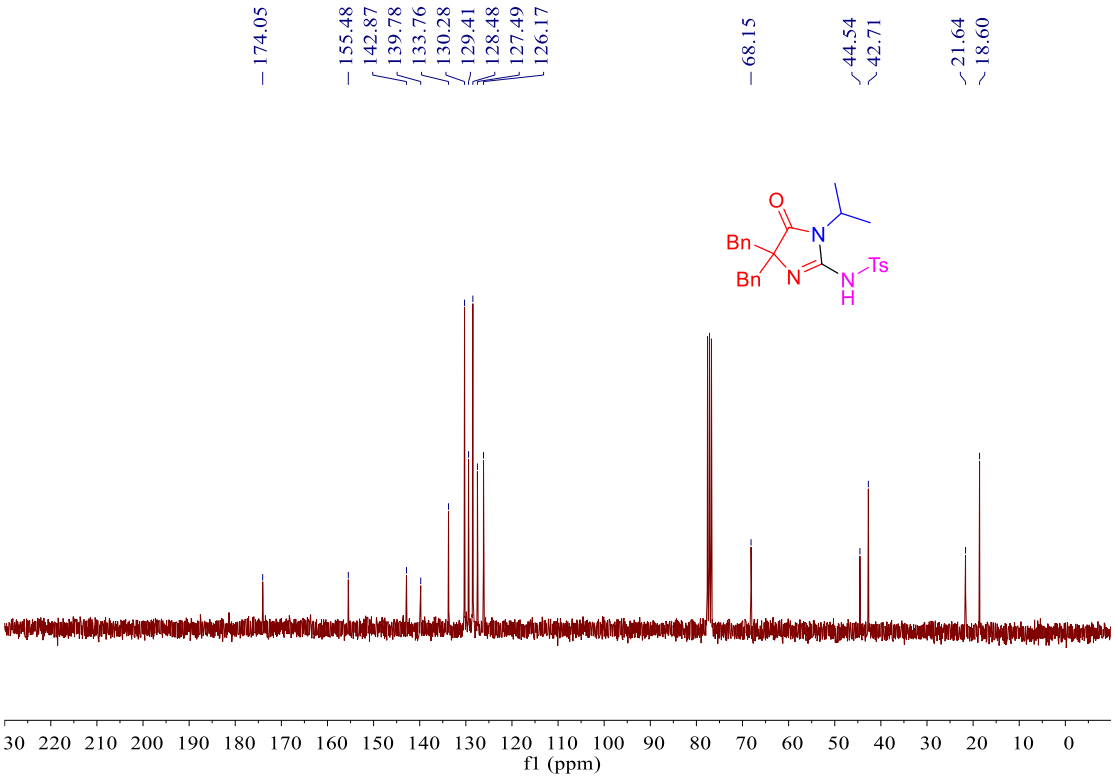
^{13}C NMR Spectra of **4j**



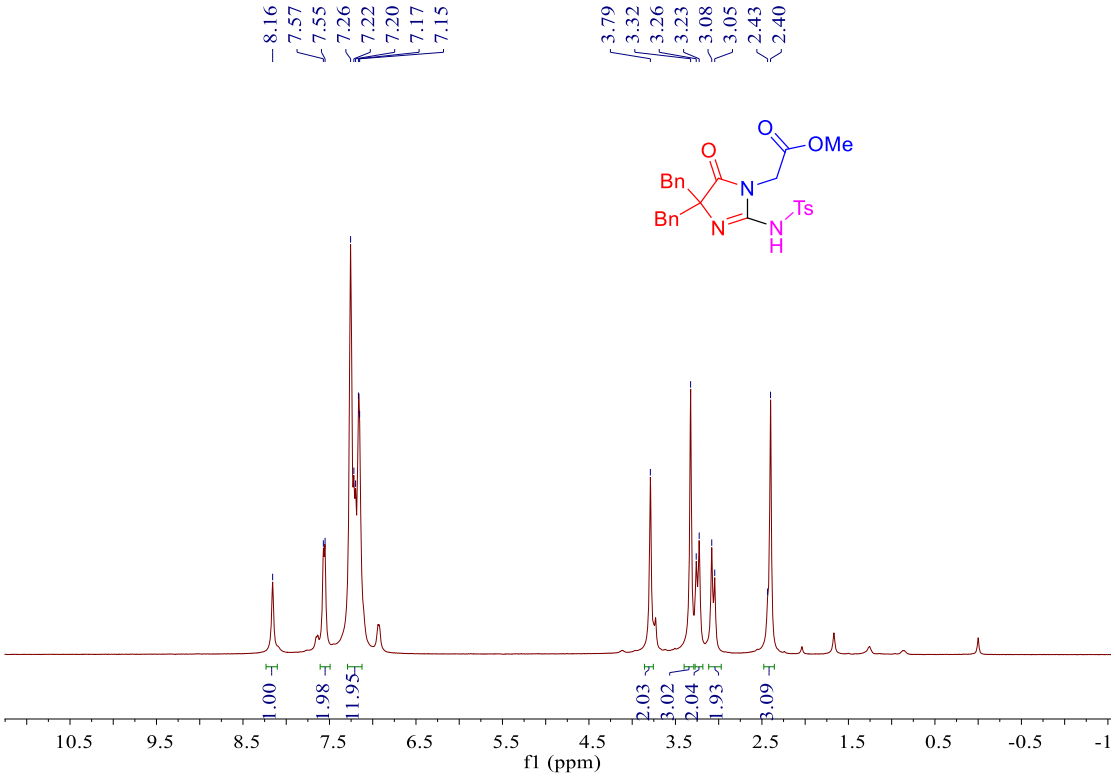
^{1}H NMR Spectra of **4k**



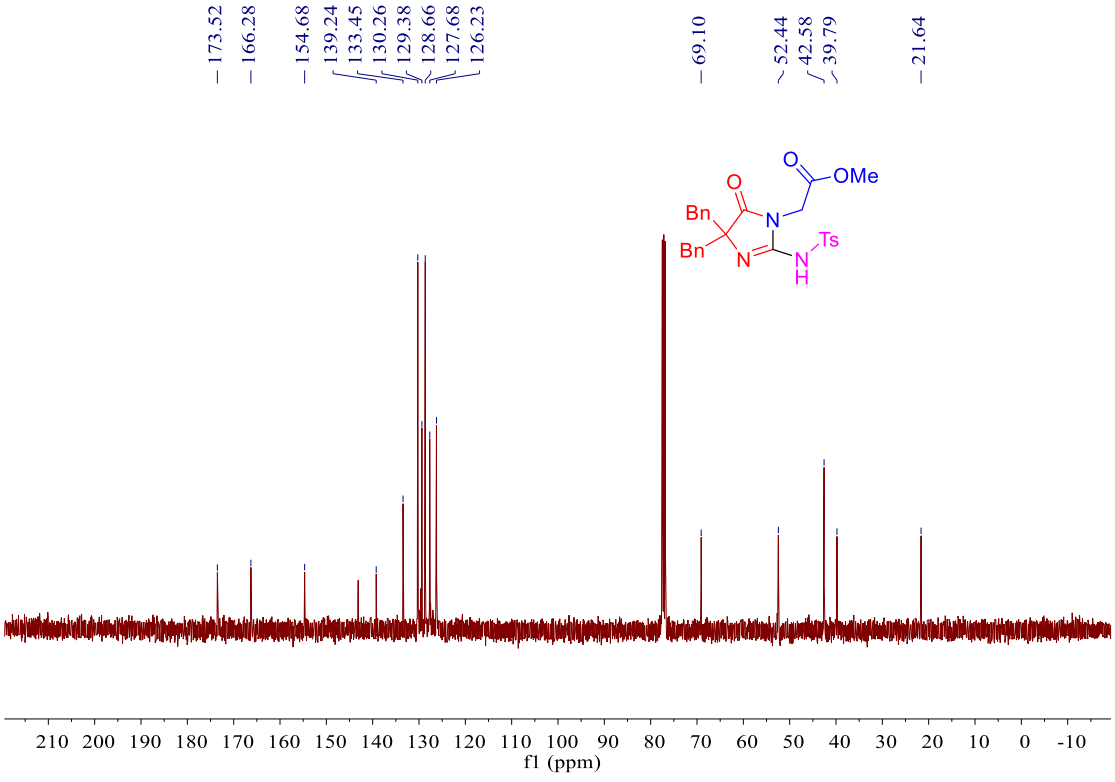
^{13}C NMR Spectra of **4k**



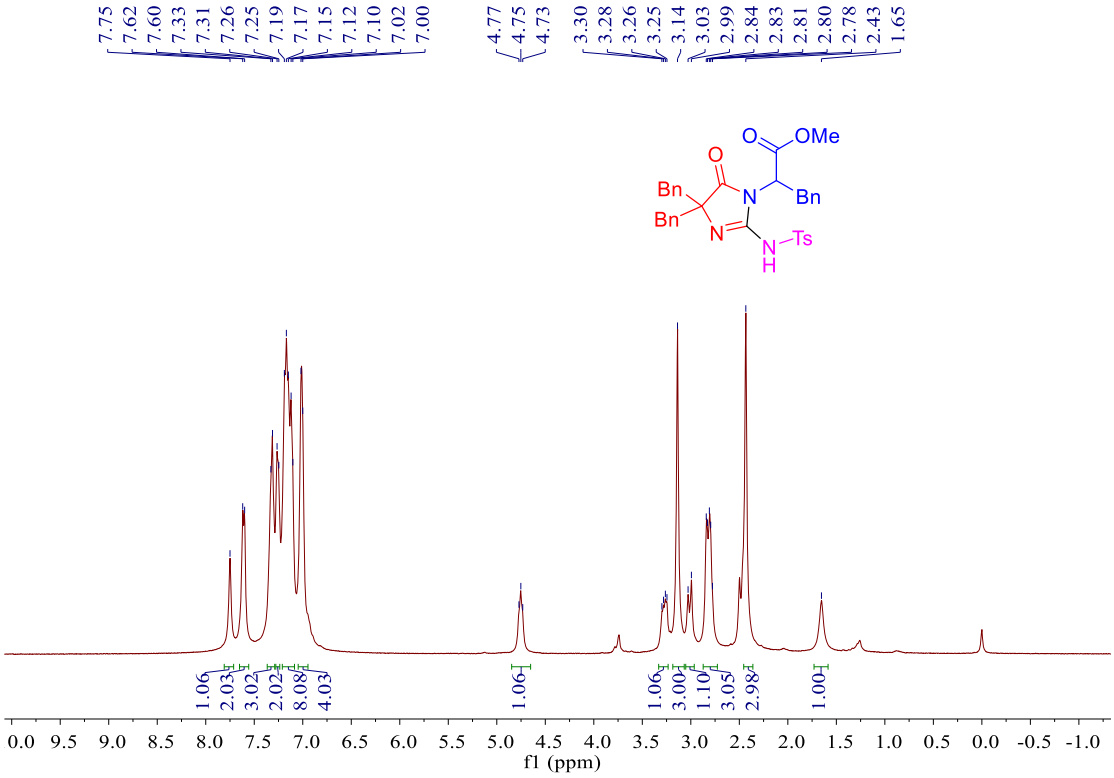
^{1}H NMR Spectra of **4l**



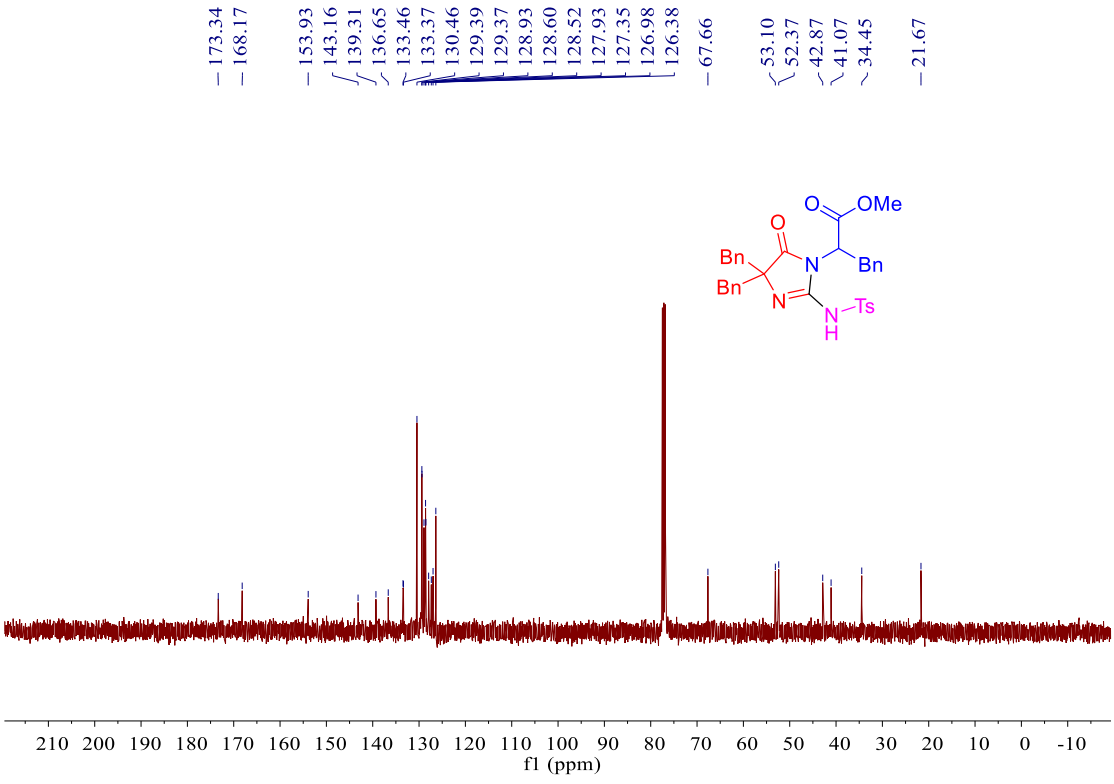
^{13}C NMR Spectra of **4l**



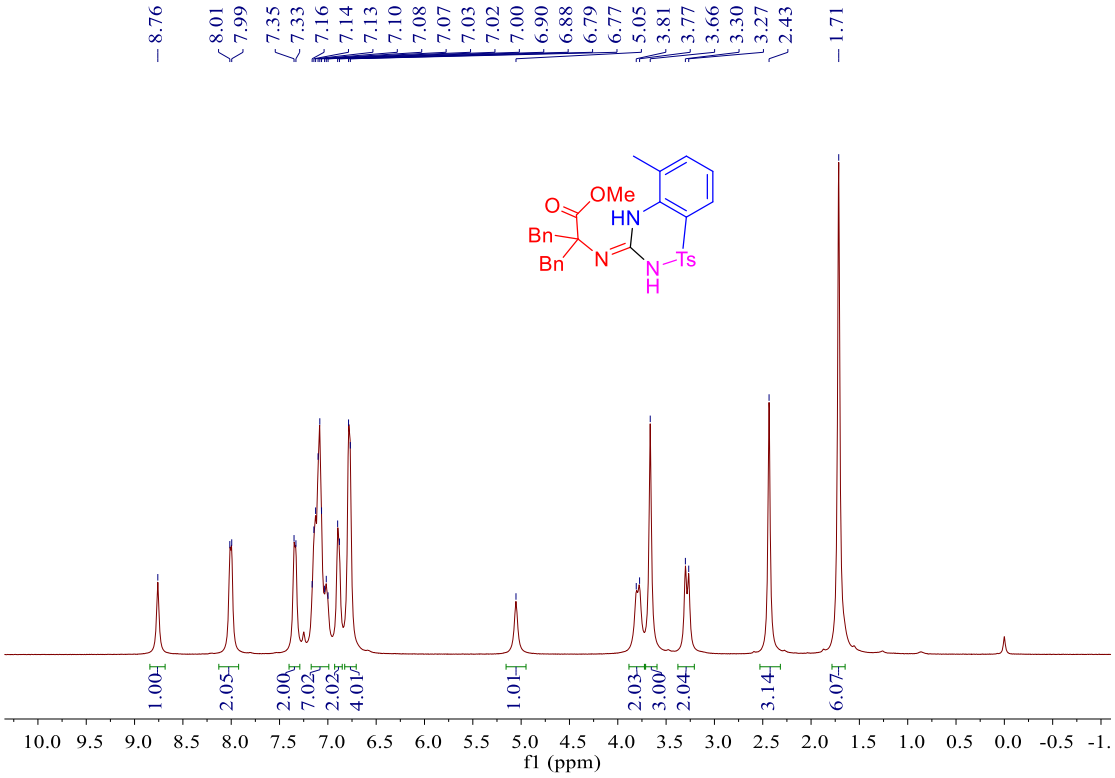
^{1}H NMR Spectra of **4m**



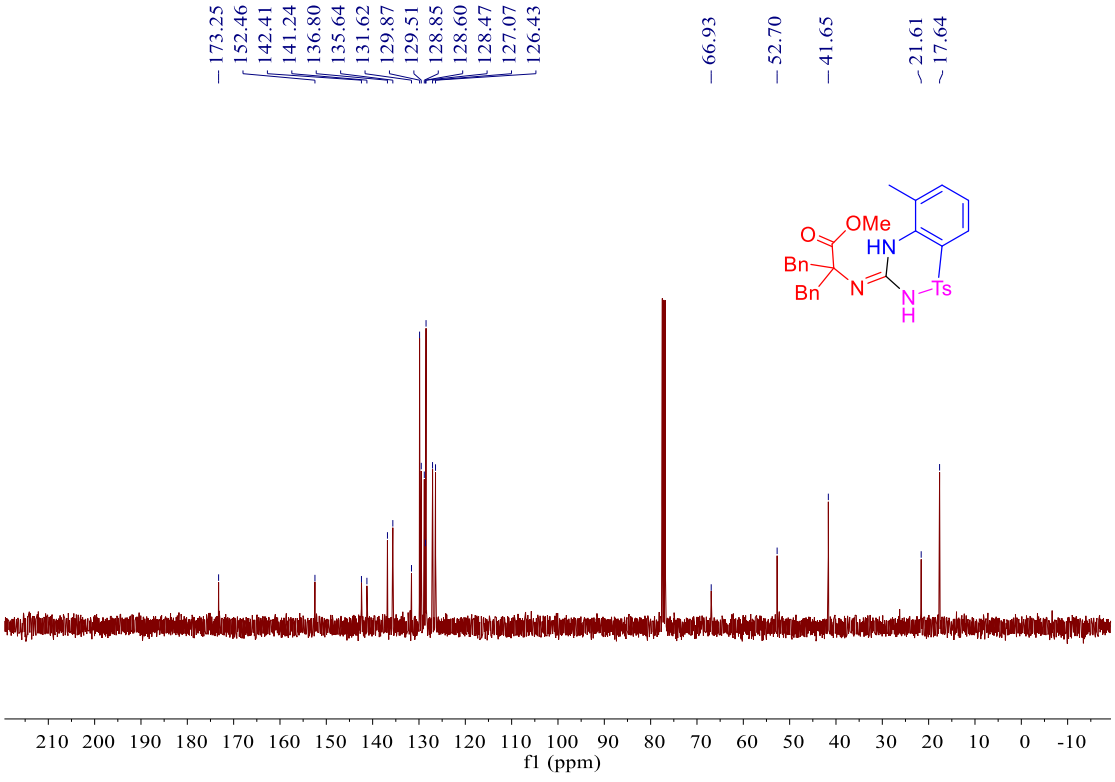
^{13}C NMR Spectra of **4m**



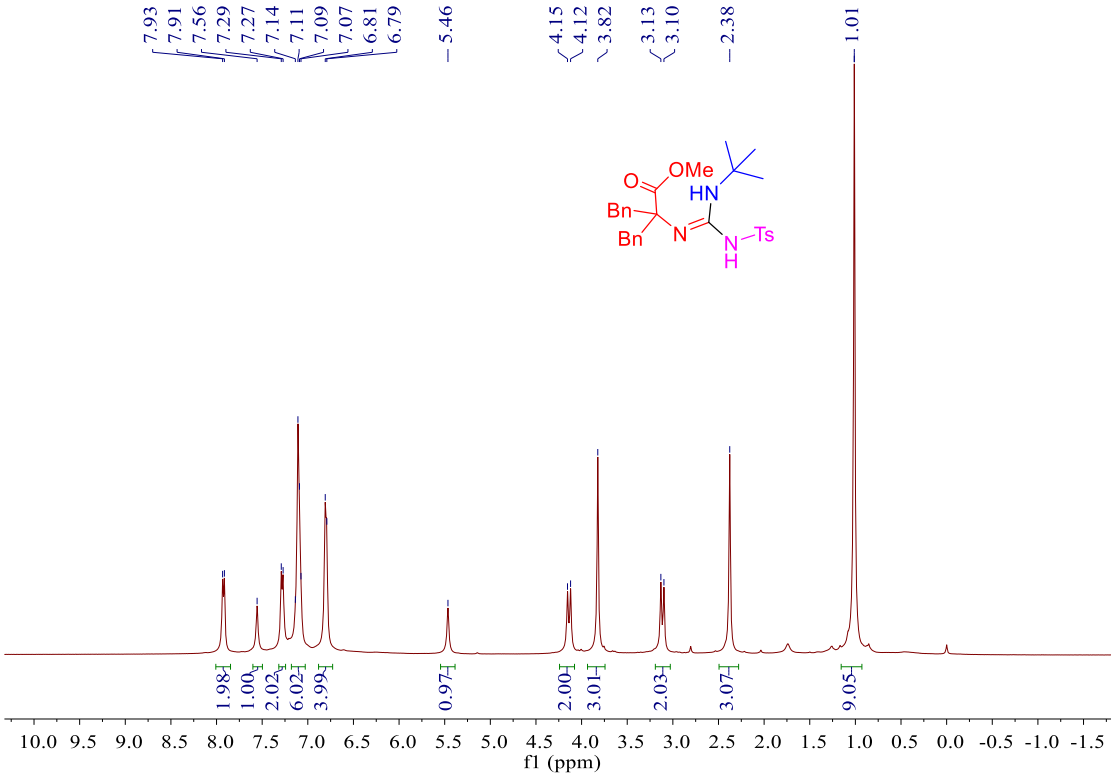
^{1}H NMR Spectra of **4n'**



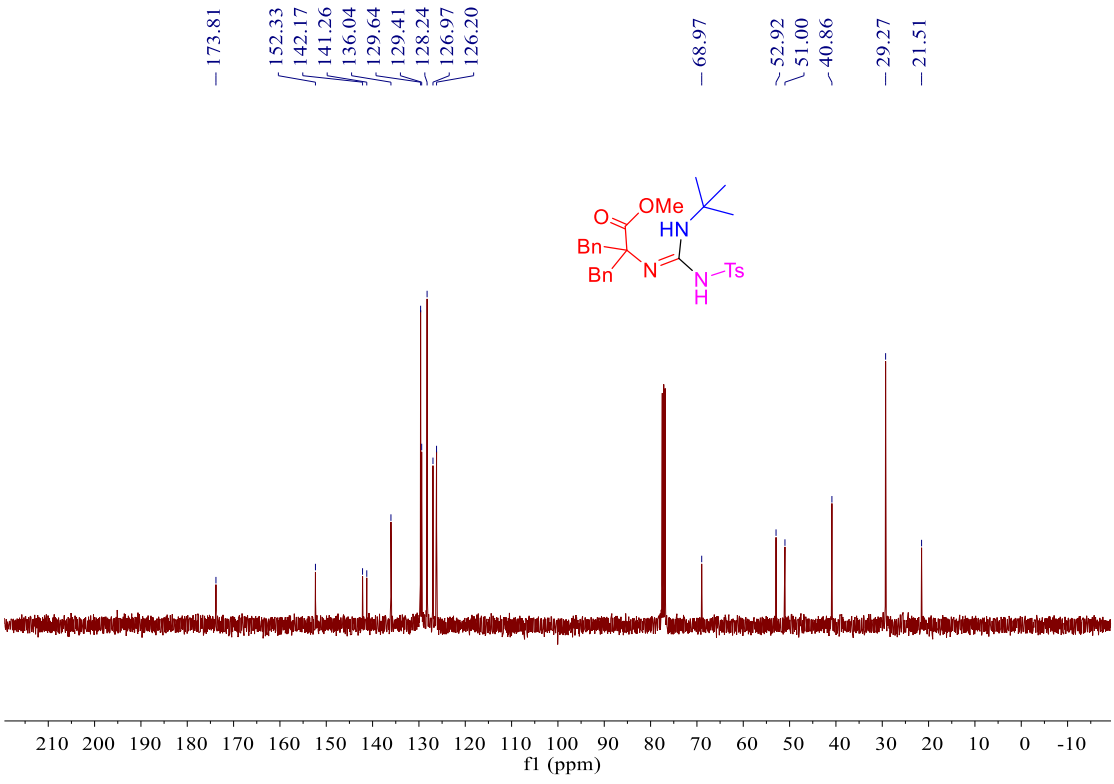
^{13}C NMR Spectra of **4n'**



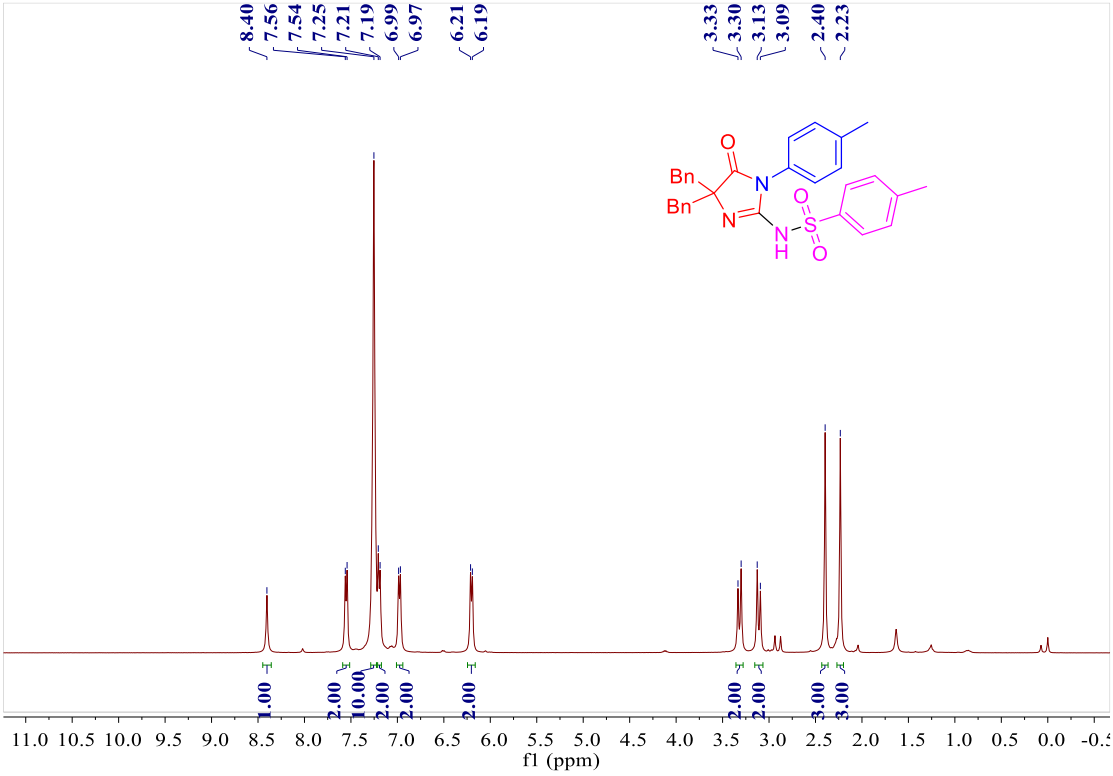
^{1}H NMR Spectra of **4o'**



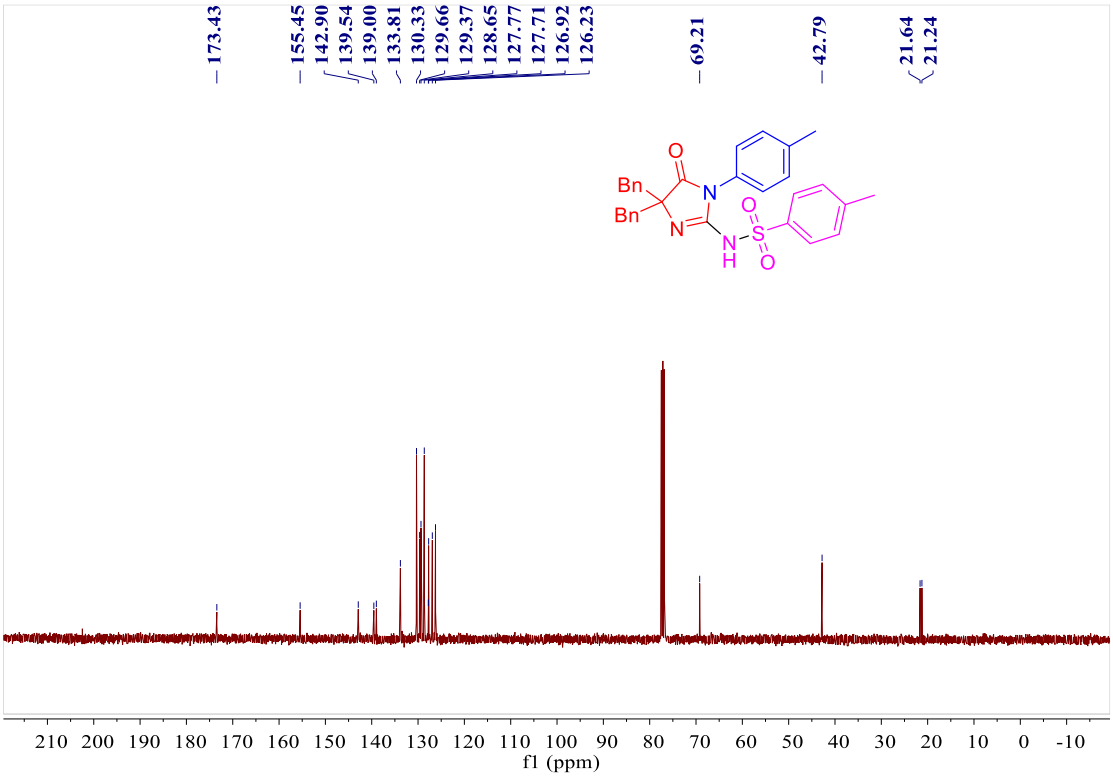
^{13}C NMR Spectra of **4o'**



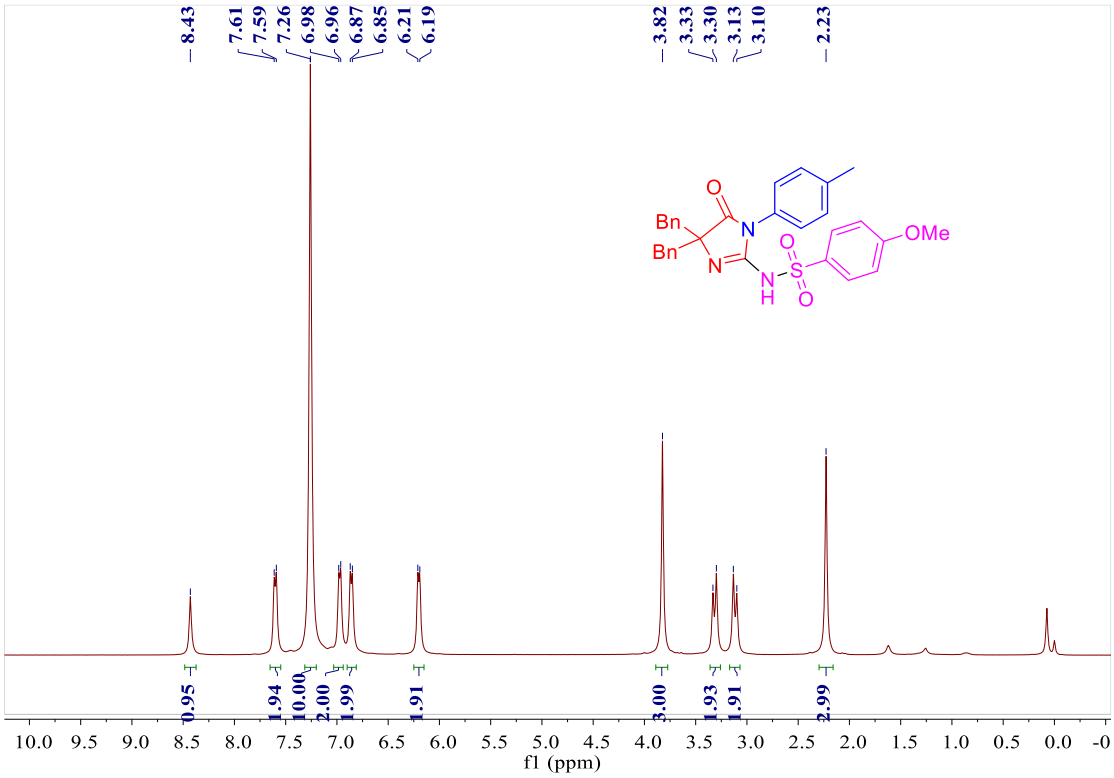
^{1}H NMR Spectra of **5a**



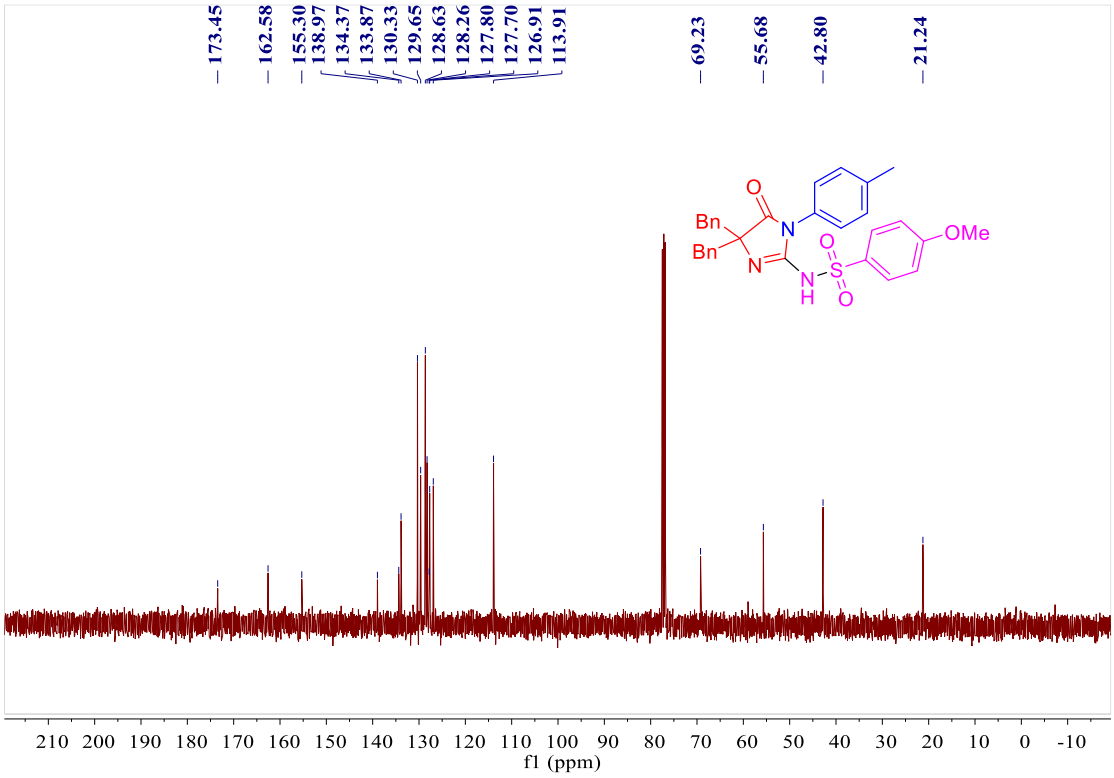
^{13}C NMR Spectra of **5a**



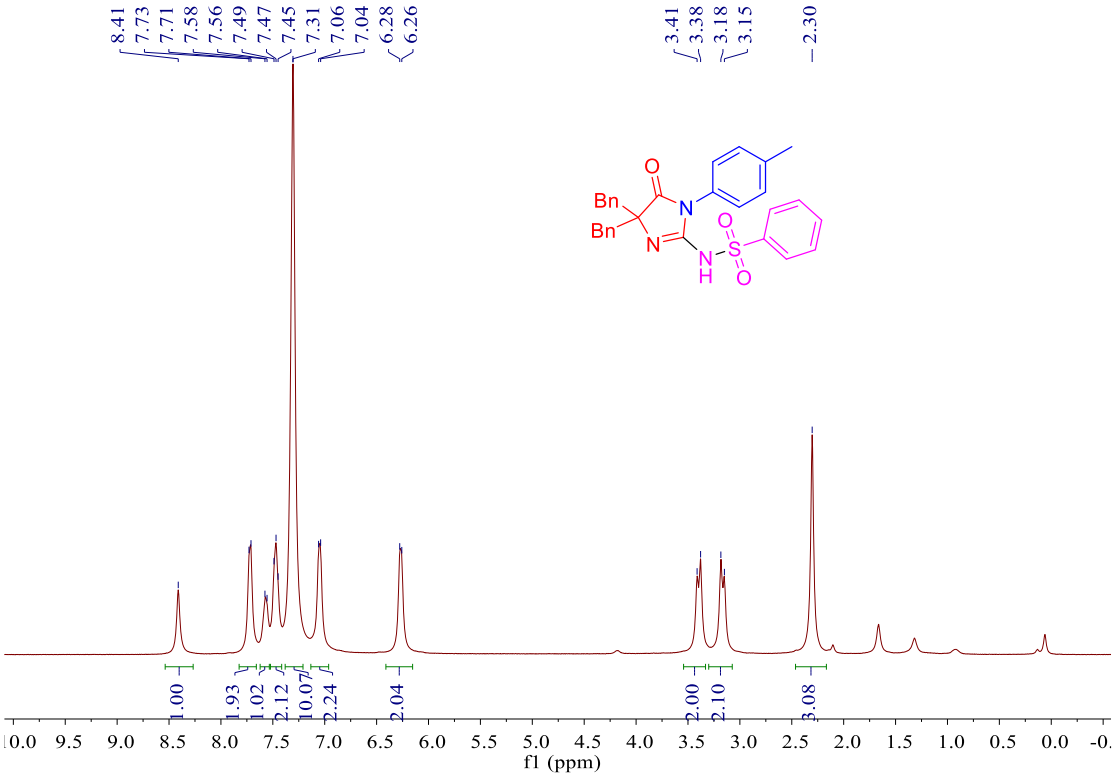
^{1}H NMR Spectra of **5b**



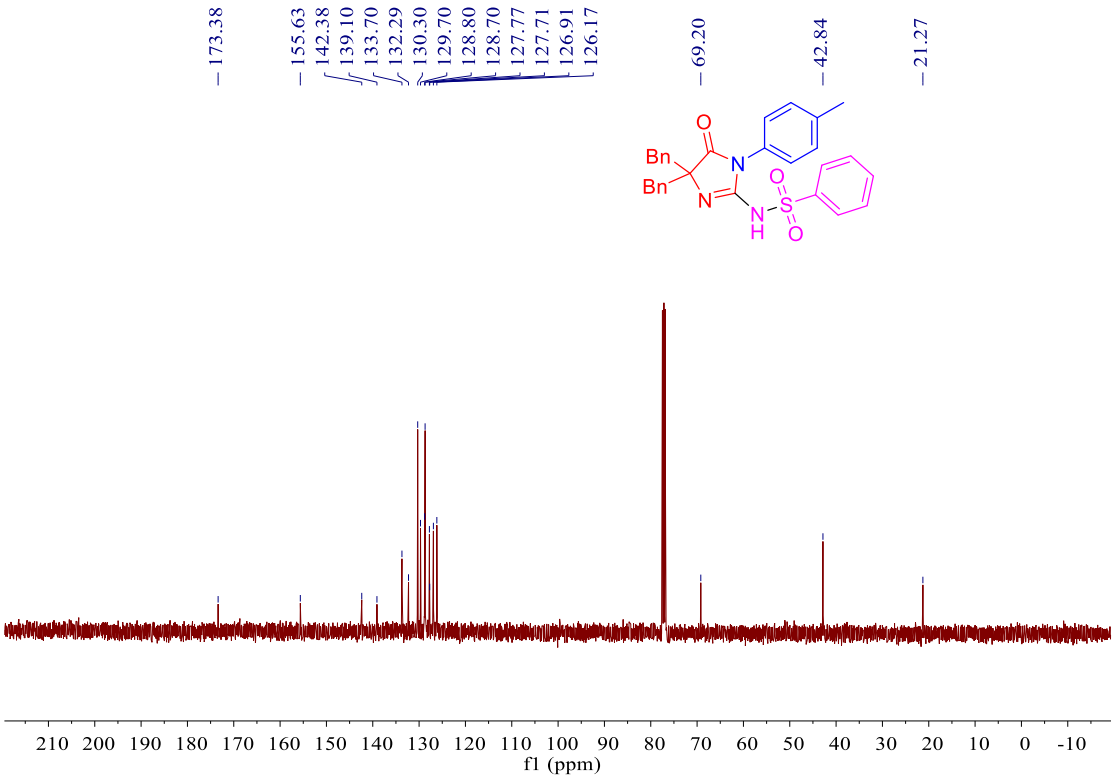
^{13}C NMR Spectra of **5b**



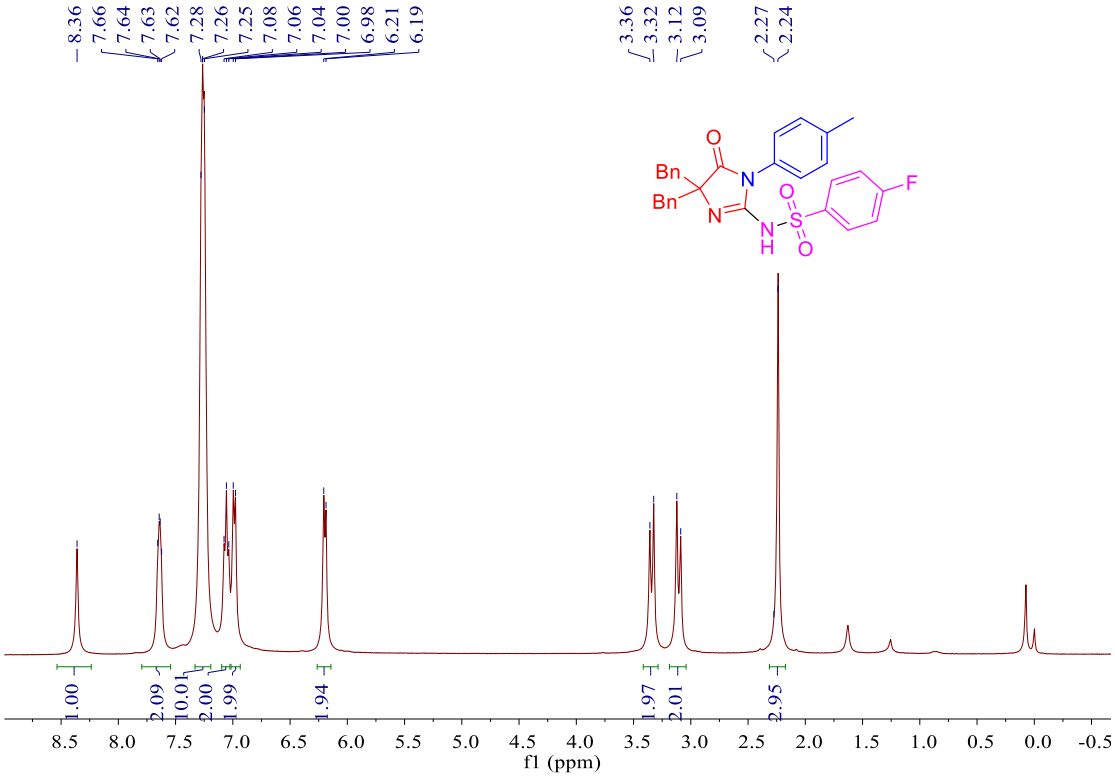
^{1}H NMR Spectra of **5c**



^{13}C NMR Spectra of **5c**



^{1}H NMR Spectra of **5d**



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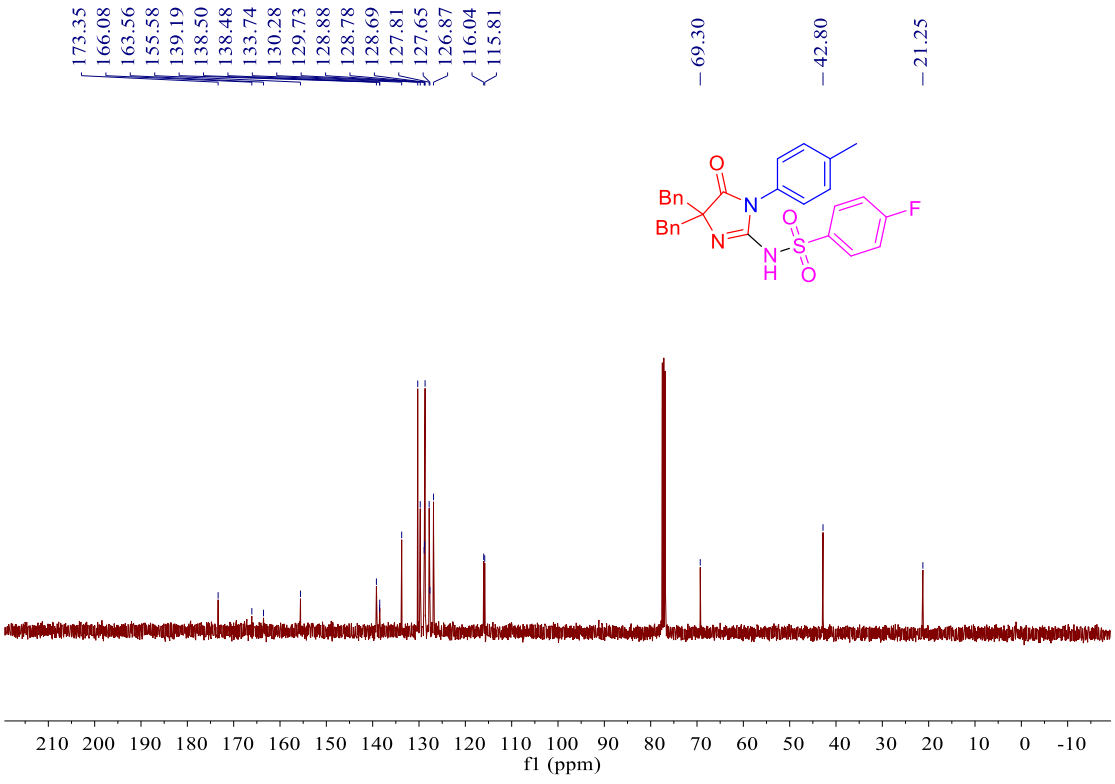

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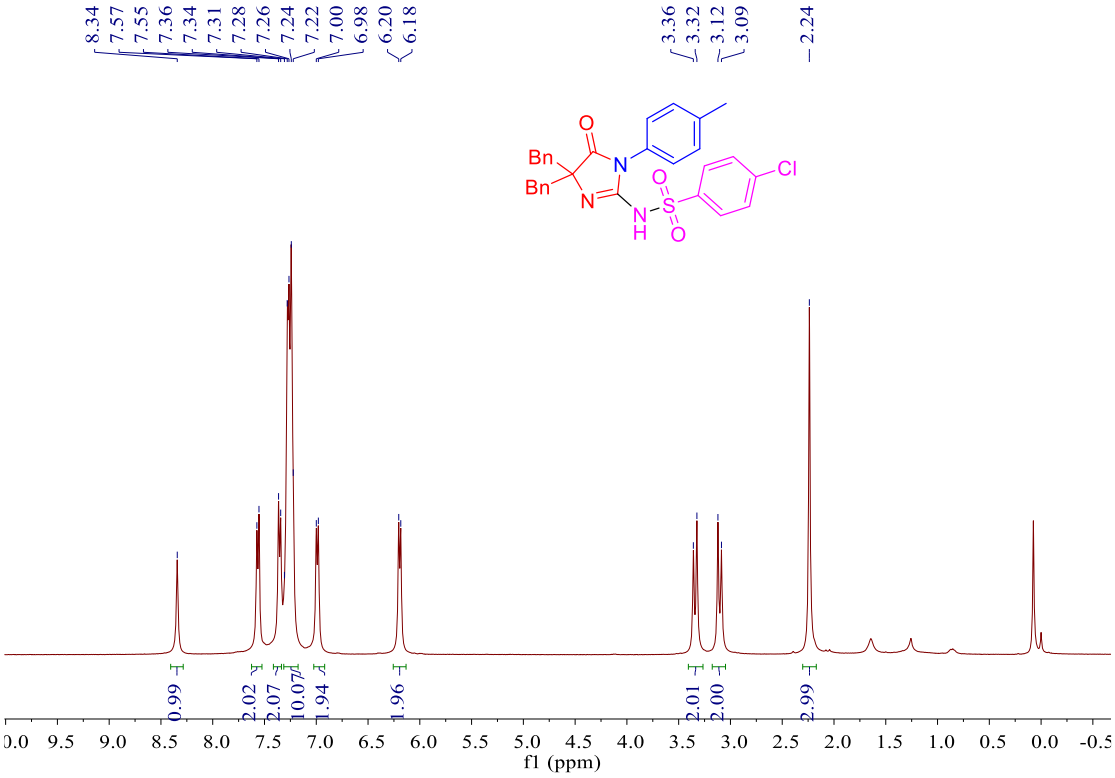

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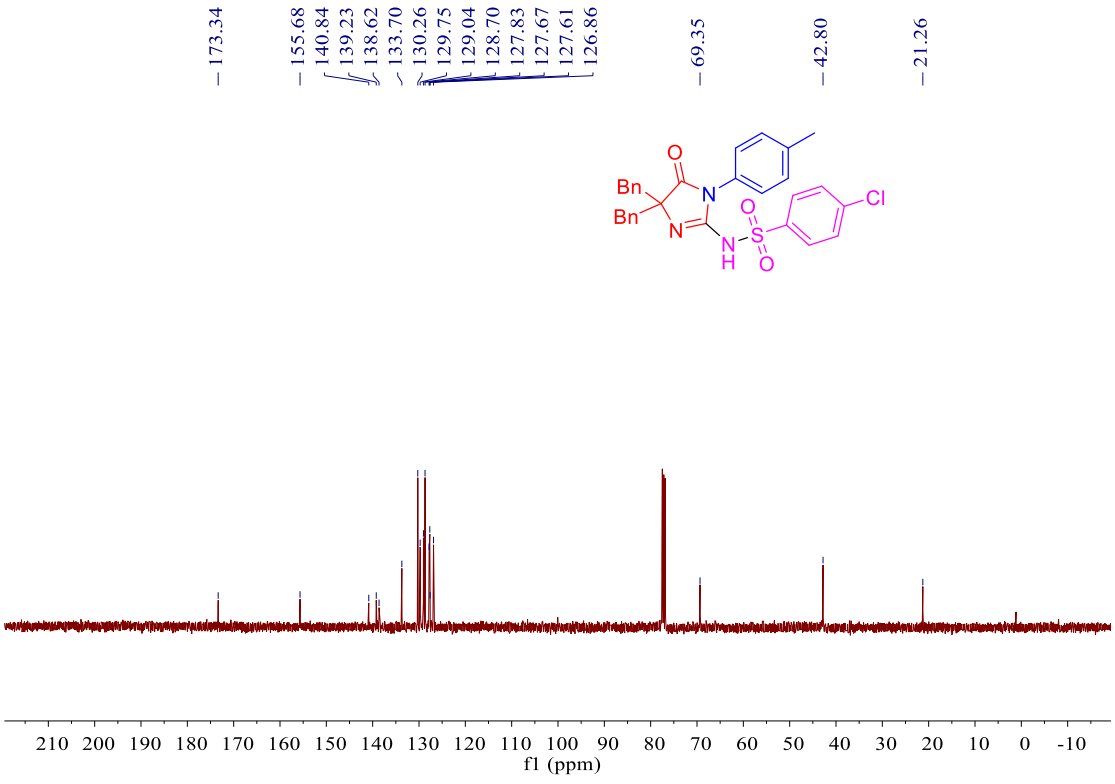
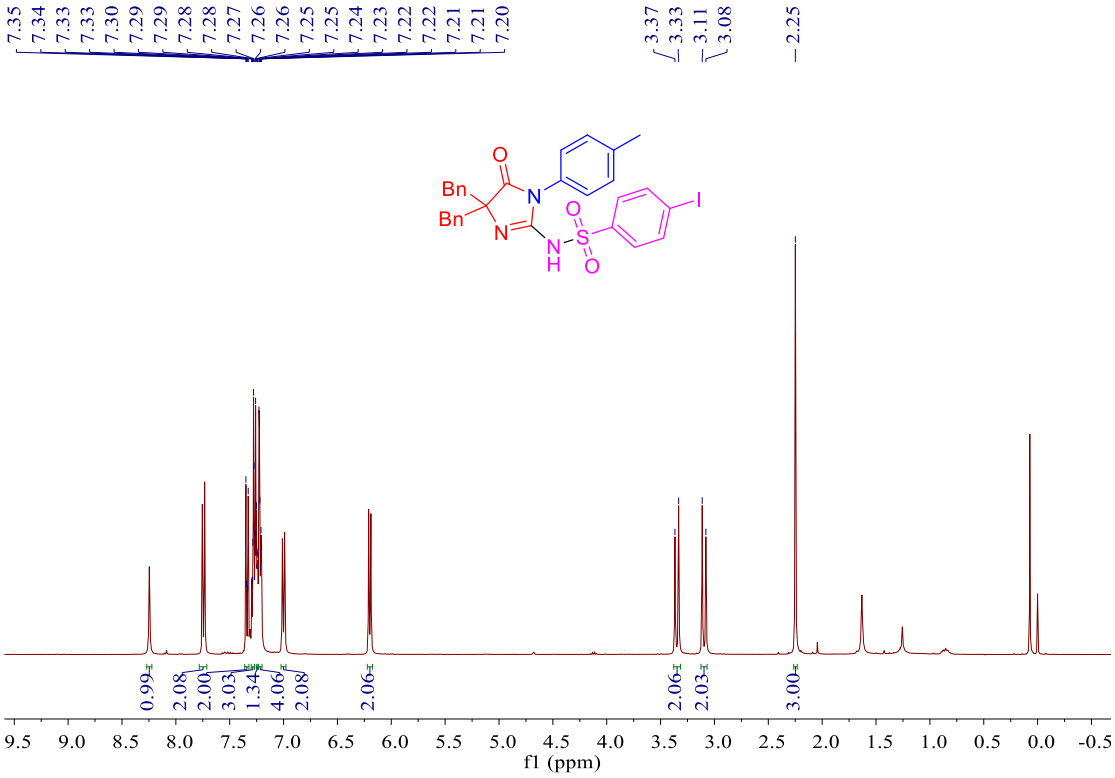


^{1}H NMR Spectra of **5f**



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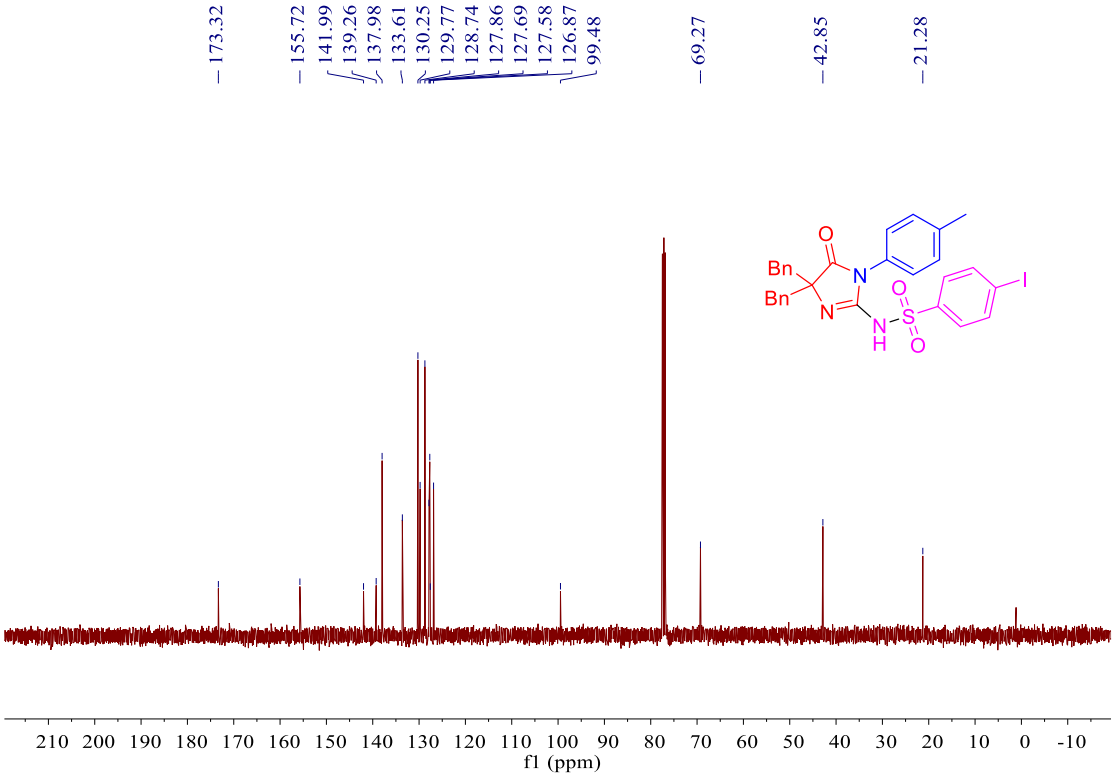
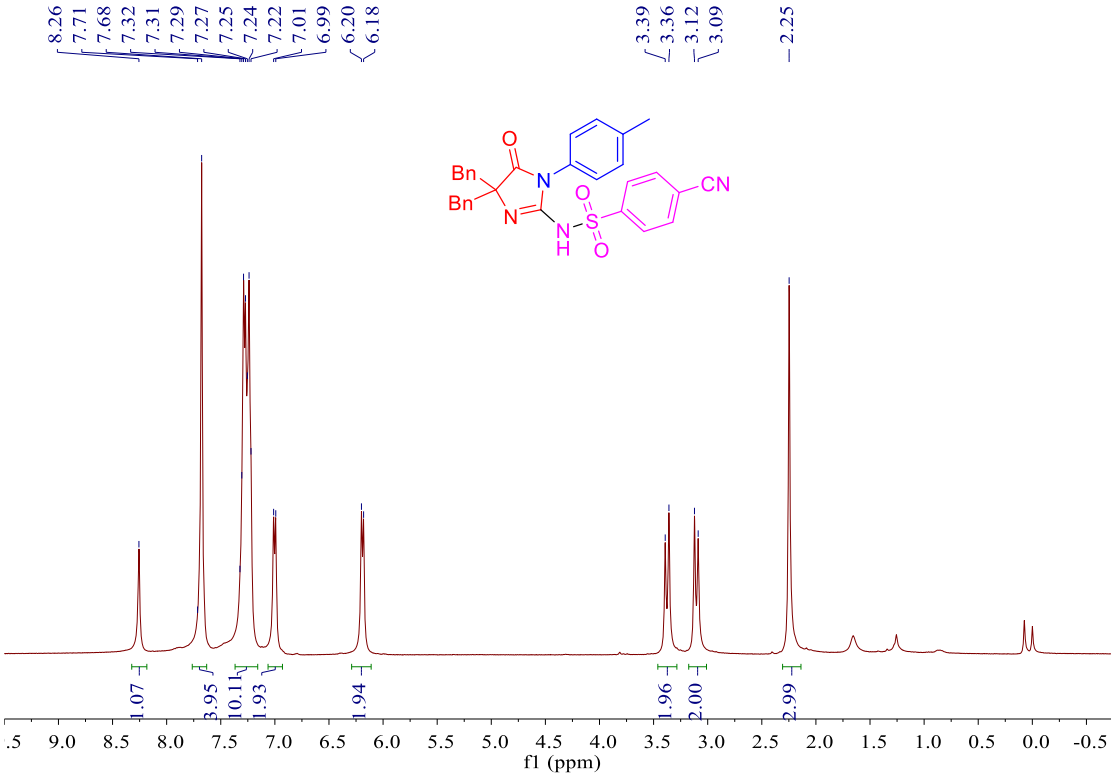
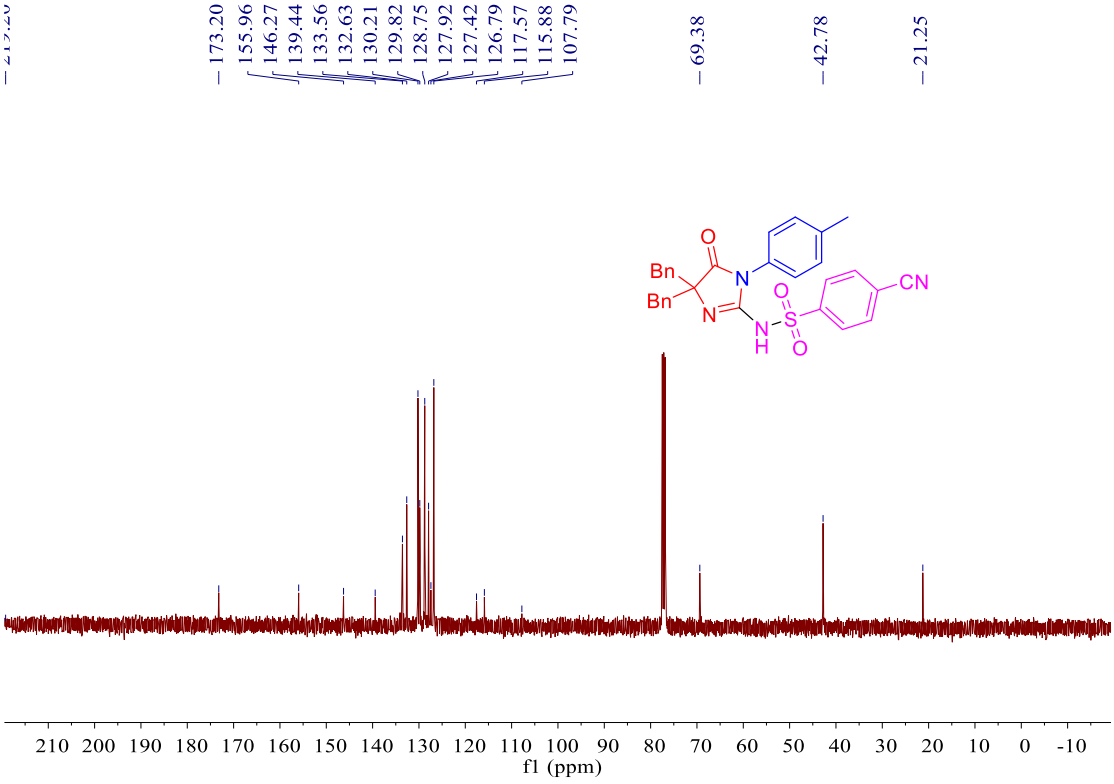


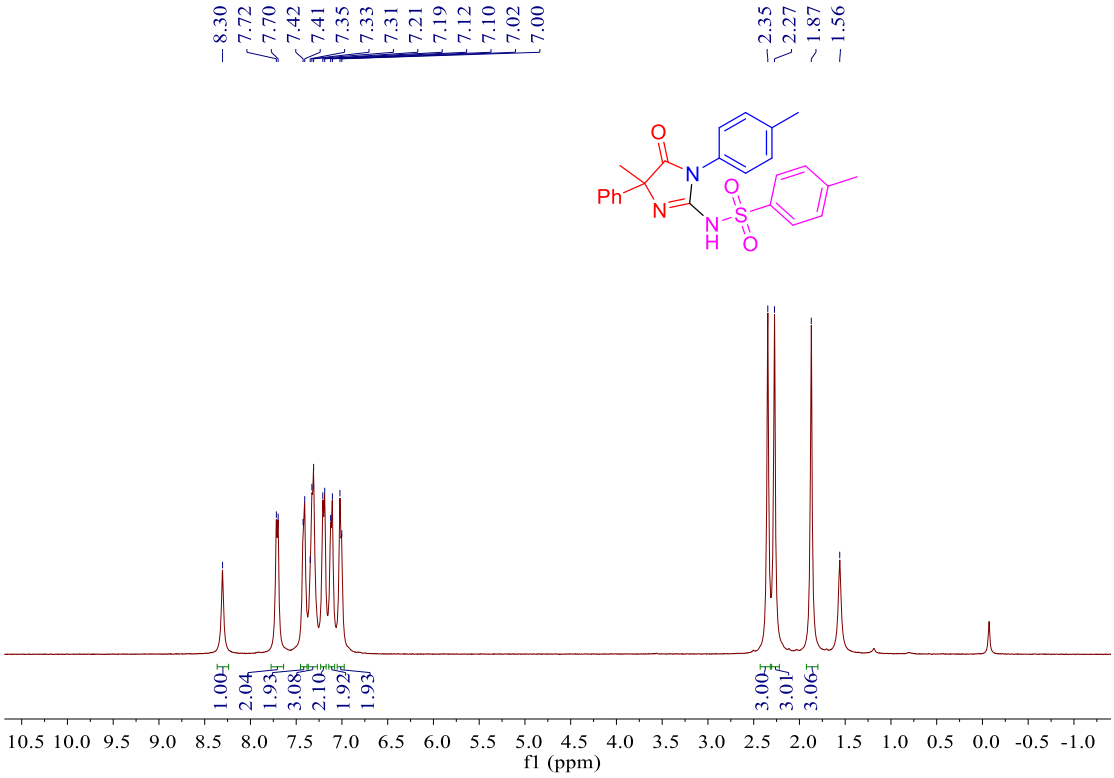
^{1}H NMR Spectra of **5g**



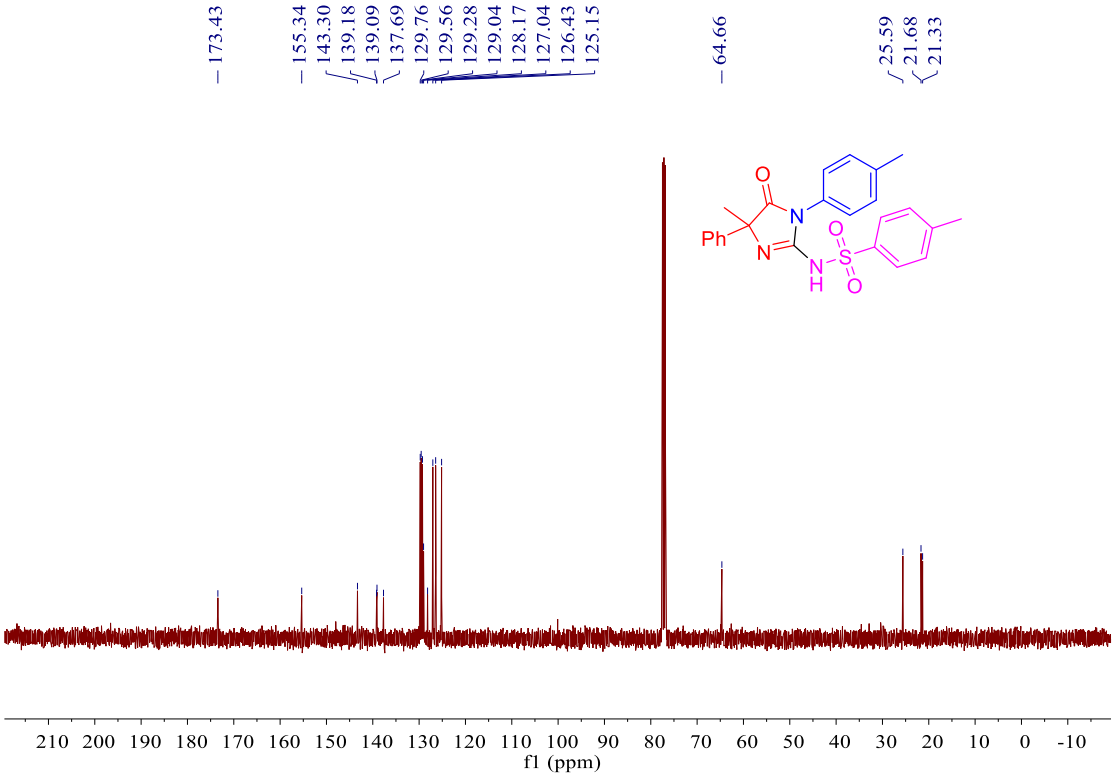
^{13}C NMR Spectra of **5g**



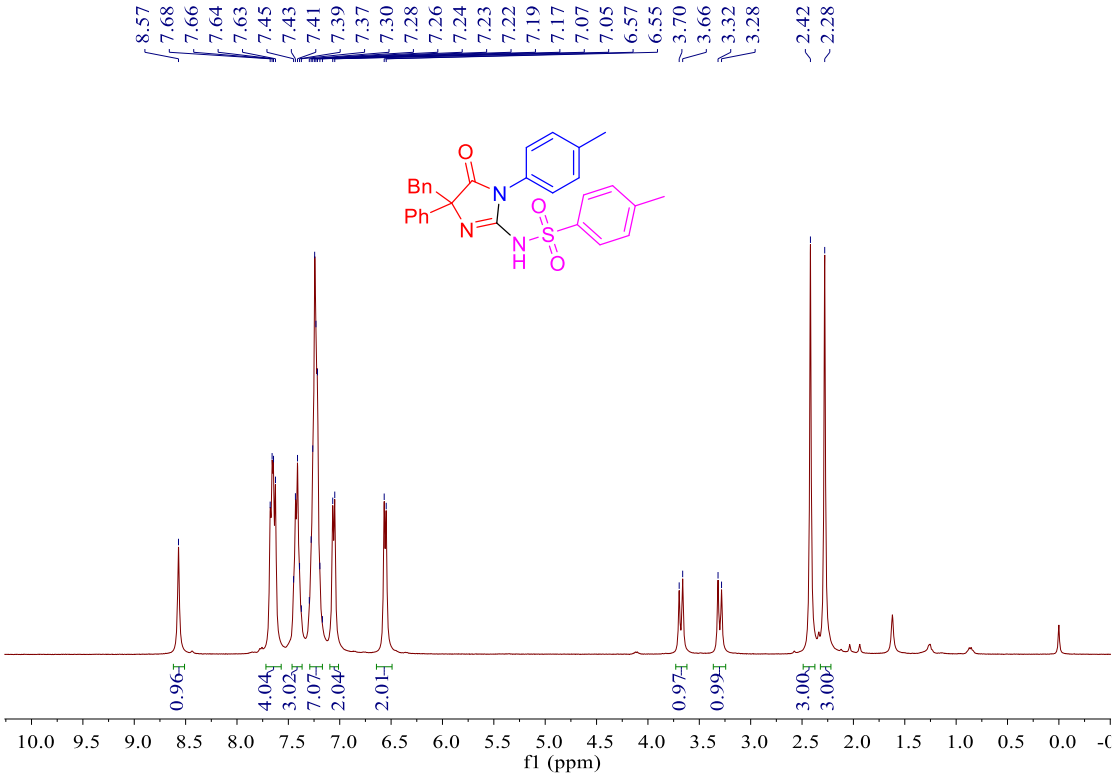
^{1}H NMR Spectra of **6d**



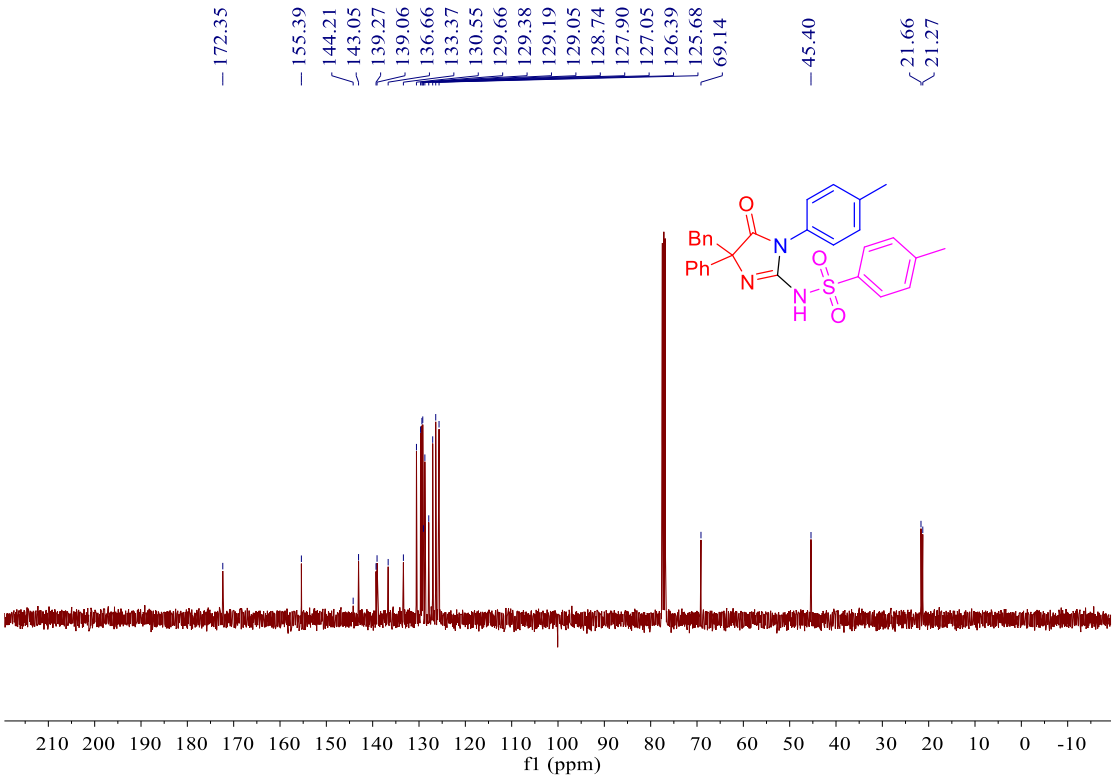
^{13}C NMR Spectra of **6d**



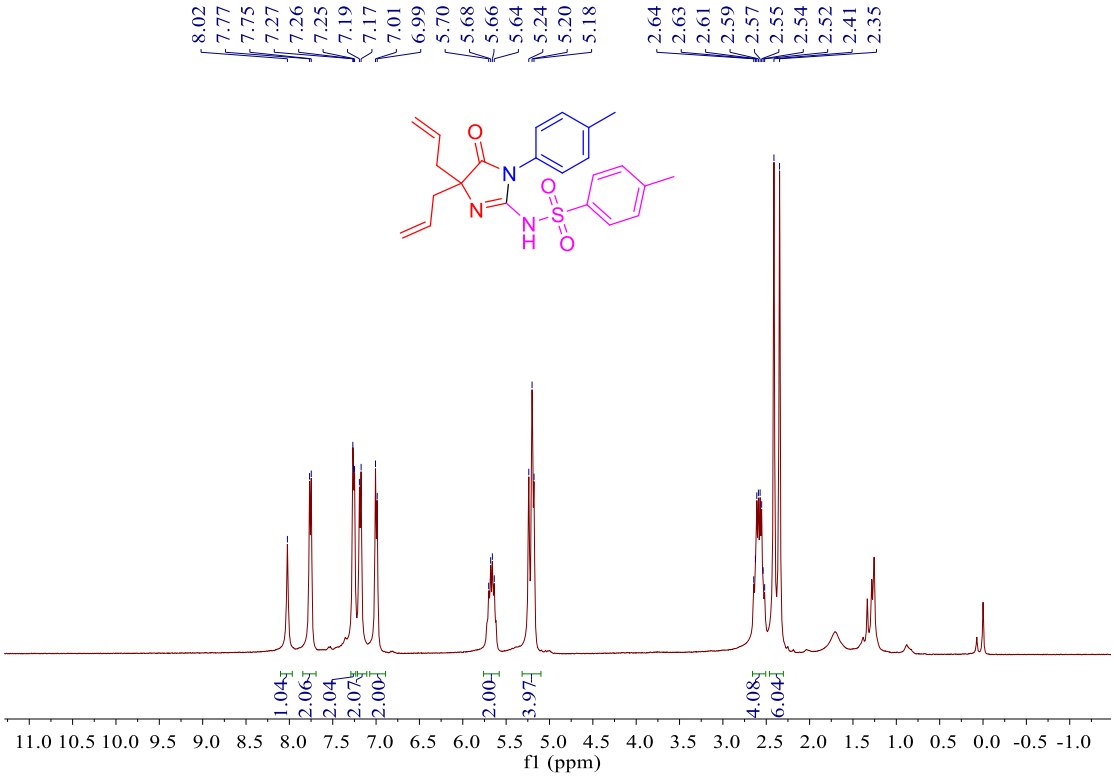
^{1}H NMR Spectra of **6e**



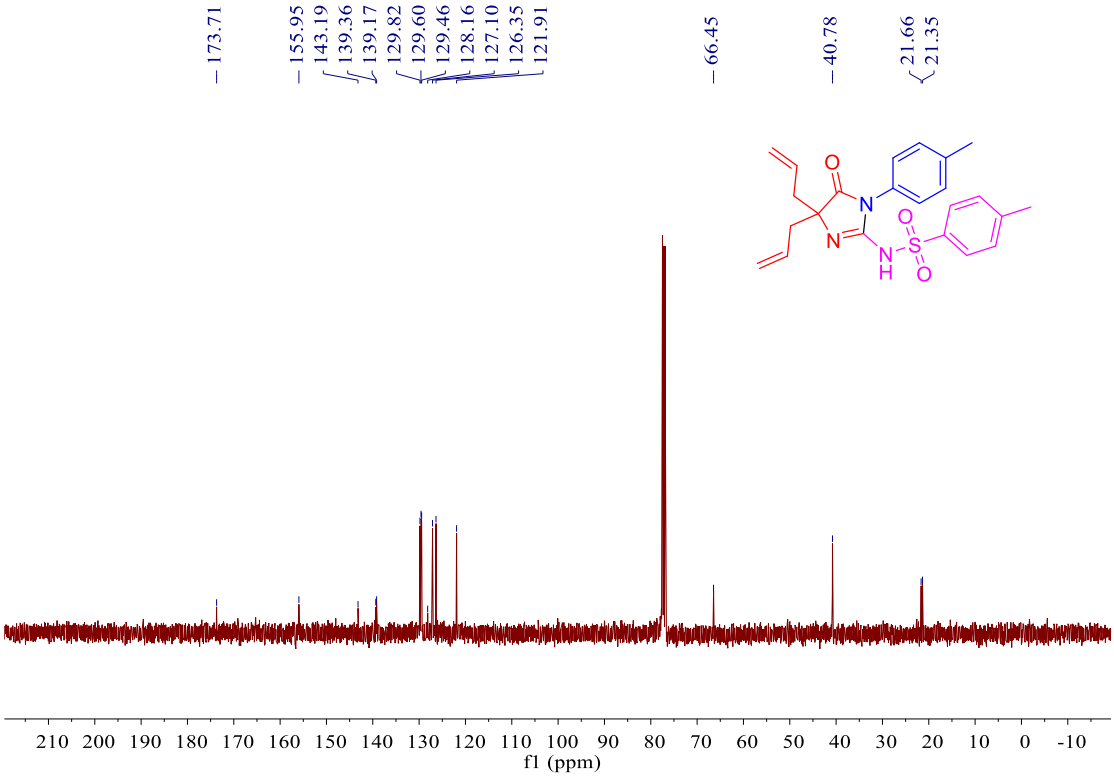
^{13}C NMR Spectra of **6e**



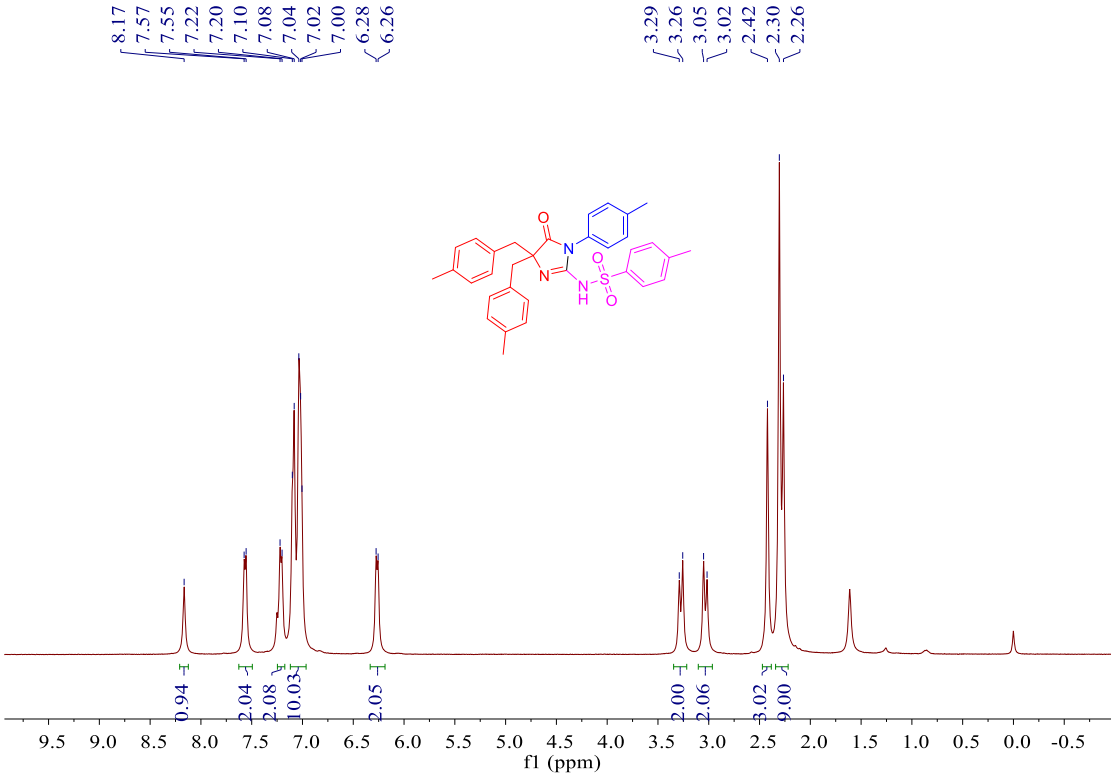
^{1}H NMR Spectra of **6f**



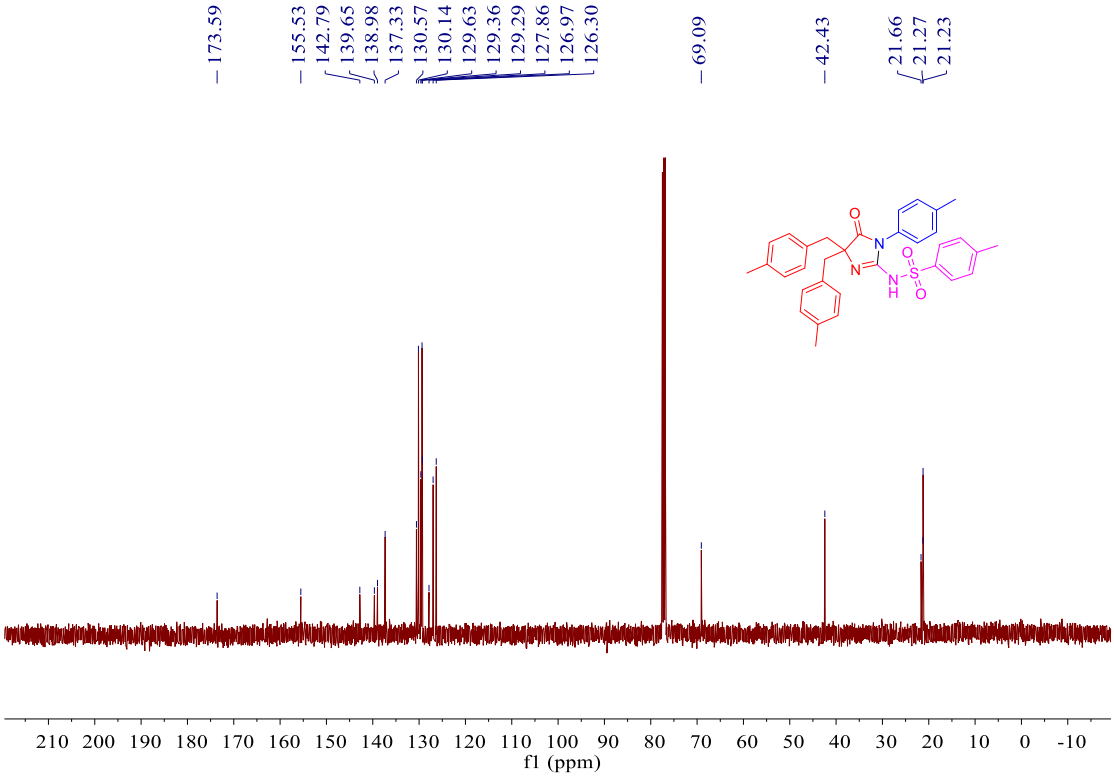
^{13}C NMR Spectra of **6f**



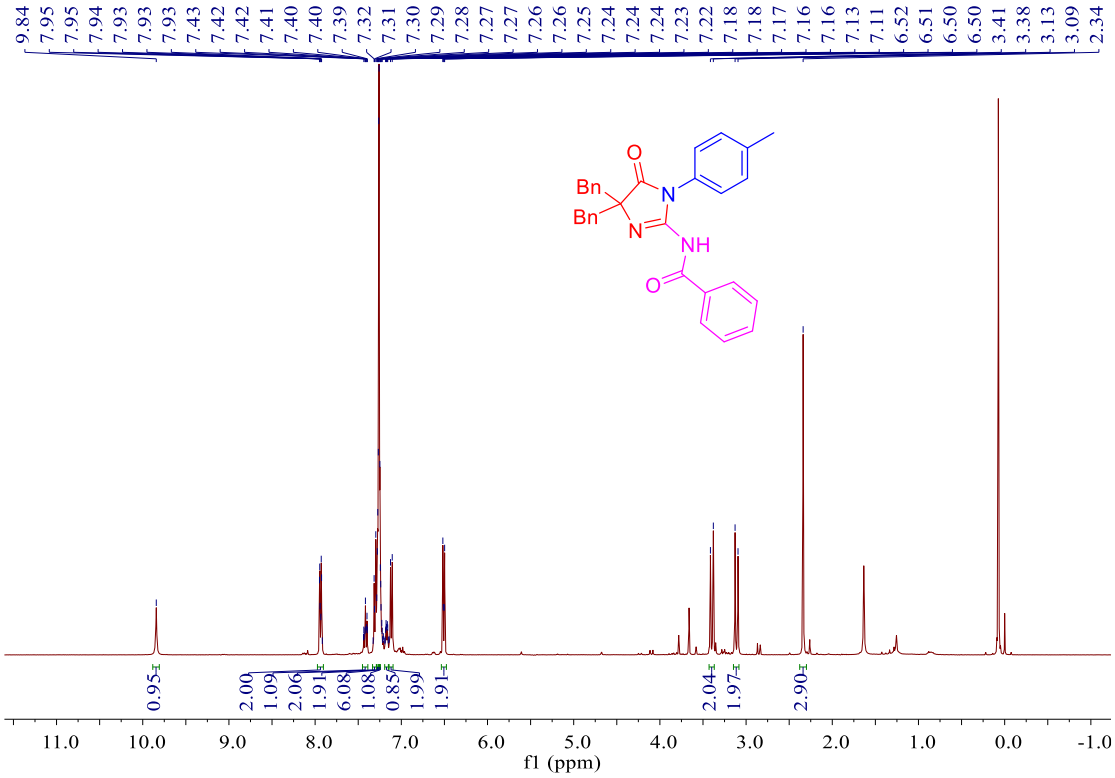
^{1}H NMR Spectra of **6g**



^{13}C NMR Spectra of **6g**



^{1}H NMR Spectra of **7a**



^{13}C NMR Spectra of **5j**