Electronic Supplementary Information

Ring expansion and fused cyclization catalysis to construct indoloquinazolinones with functionalization

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1.Materials and Methods

Unless otherwise stated, reactions were performed in oven-dried glassware fitted with rubber septa and were stirred with teflon-coated magnetic stirring bars. Liquid reagents and solvents were transferred via syringe using standard Schlenk techniques. All the solvents and reagents were used as received unless otherwise noted. Petroleum ether used in our experiments was in the boiling range of 60-80 °C. Reaction temperatures above 25 °C refer to oil bath temperature. Thin layer chromatography was performed using silica gel 60 F-254 precoated plates (0.25 mm) and visualized by UV irradiation, anisaldehyde stain and other stains. Silica gel of particle size 100-200 mesh was used for column chromatography. Melting points are recorded on a digital melting point apparatus and are uncorrected. ^{1}H and ^{13}C NMR spectra were recorded in a 300 MHz and 400 MHz spectrometers with ^{13}C operating frequencies of 75 MHz and 100MHz, respectively. Chemical shifts (δ) are reported in ppm relative to the residual solvent $CDCl_3$ signal (δ = 7.24 for ^{1}H NMR and δ = 77.0 for ^{13}C NMR), DMSO-d6 signal (δ = 2.47 for ^{1}H NMR and δ = 39.4-40.6 for ^{13}C NMR) and CD_3OD signal (δ = 49.0 for ^{13}C NMR). Data for ^{1}H NMR spectra are reported as follows: chemical shift (multiplicity, number of hydrogen and coupling constants). Abbreviations are as follows: s (singlet), d (doublet), t (triplet), q (quartet), m (multiplet),br (broad). IR spectra were recorded on a FT-IR system and are reported in frequency of absorption (cm^{-1}). Only selected IR absorbance is reported. High-Resolution Mass Spectrometry (HRMS) data was recorded on a Qtof-micro quadruple mass spectrophotometer using acetonitrile as a solvent.

2. Isatins Used in this Reaction(1a-g)

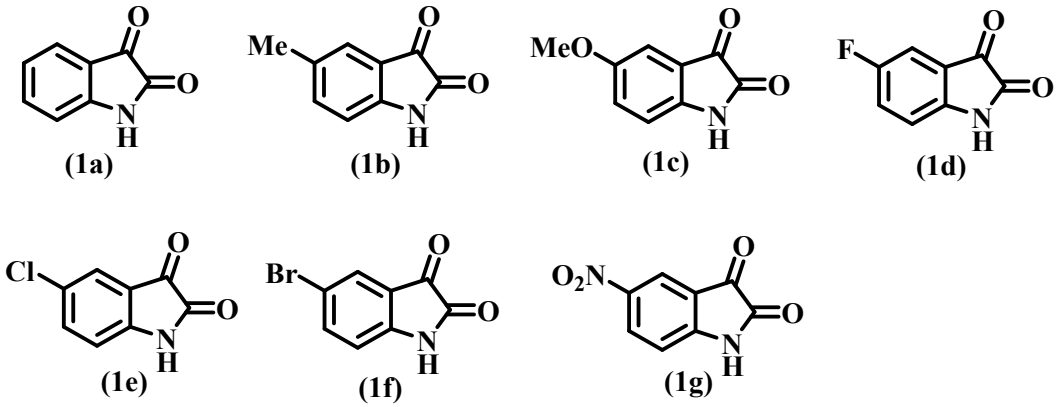
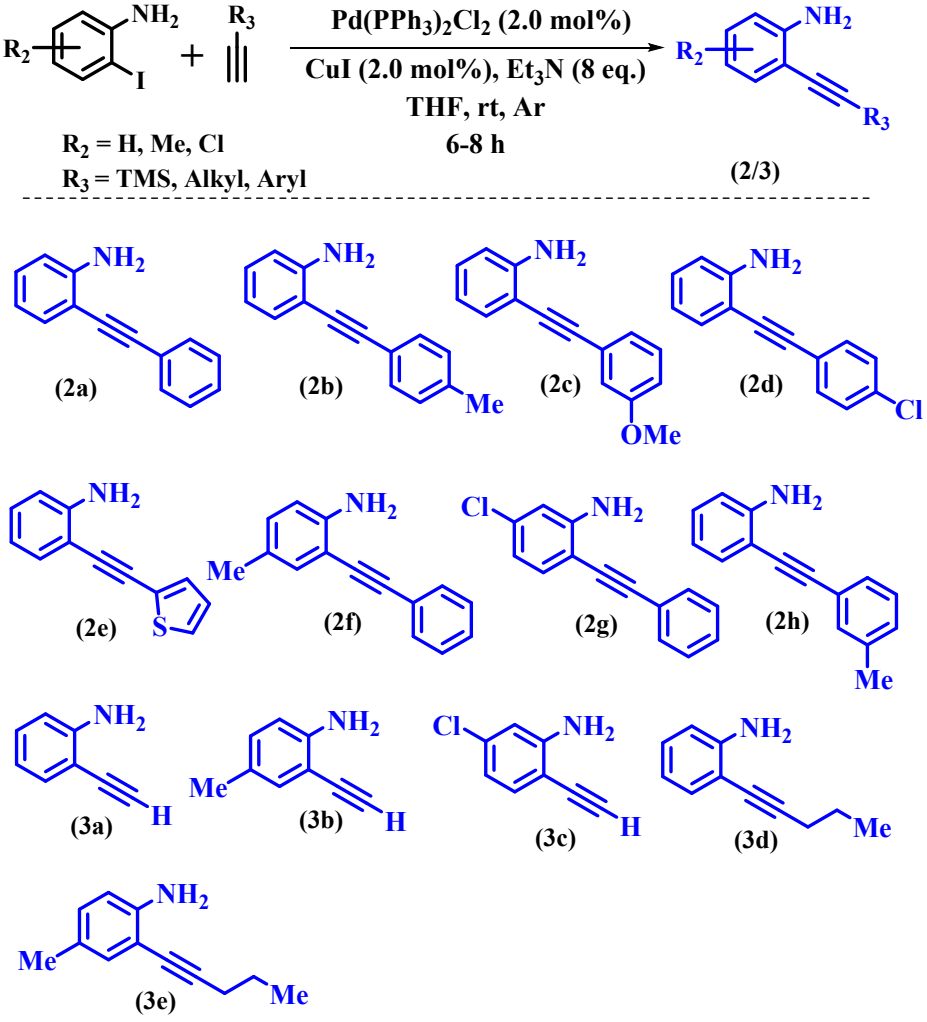


Figure S1. Isatins used in this reactions(**1a-g**)

3.General Procedure for the Synthesis of *ortho*-Alkynylanilines (2a-g and 3a-f)[1]**:** Catalyst CuI (0.06 mmol, 11.94 mg),$Pd(PPh_3)_2Cl_2$ (0.06 mmol, 42.06 mg) and Et_3N (24 mmol, 3.3 ml) were added to a well stirred solution of 2-iodo aniline (3 mmol) in THF (10 ml). The reaction mixture was allowed to stir at room temperature for 30 minutes, phenyl acetylene derivative (3.3 mmol,) was added drop wise, and the reaction content was stirred at room temperature for 1 h. After complete consumption of the starting material, the solvent was removed under reduced pressure, and the crude residue was purified over silica gel column chromatography to get pure 2-alkynylanilines with excellent yield.

Scheme S1. Synthesized*ortho*-alkynylanilines used in the reaction



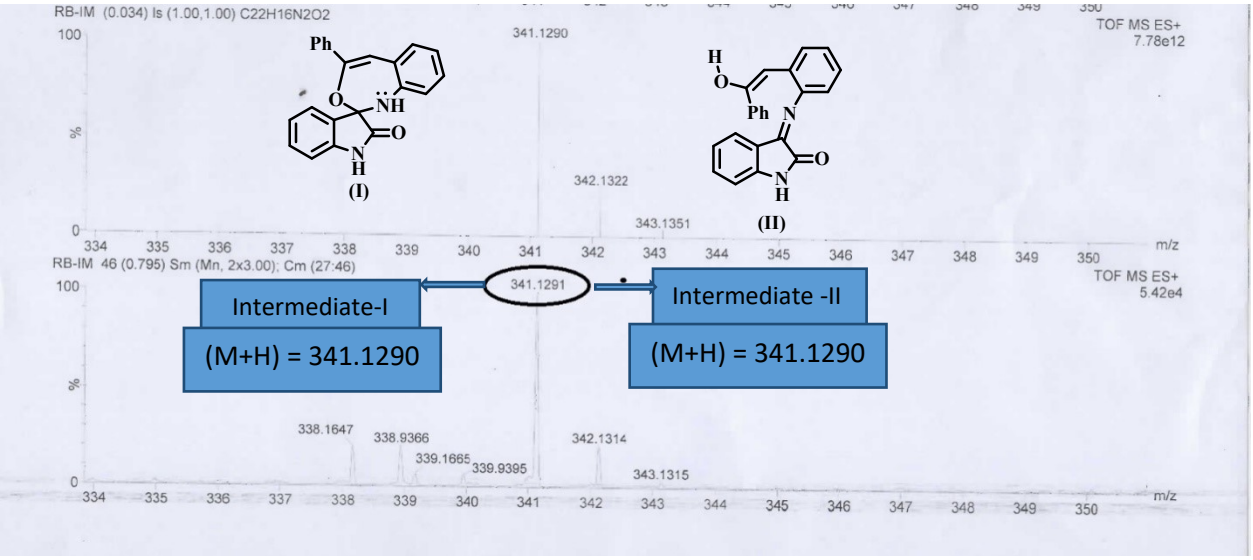
4. **General Procedure for the Synthesis of 12-Benzoylindolo[1,2-*c*]quinazolin-6(5*H*)-one derivatives [4(a-g)(a-g)]:**To a mixture of isatin(**1**, 0.50 mmol) and 2-(arylethynyl)aniline(**2**, 0.50

mmol),$FeCl_3$(24.33 mg, 0.15 mmol, 30 mol%) and$CuBr_2$(11.16 mg, .05 mmol, 10 mol%)were taken in an oven dried round bottom flask containing a teflon coated magnetic stir bar and 1,4-dioxane (3 ml) was added. The reaction mixture was set to a pre-heated oil bath having temperature 120°C and completion of the reaction was confirmed by TLC monitoring. The RB flask was picked up from oil and allowed to get room temperature. Solvent was removed in a rotary evaporator under reduced pressure and room temperature. The residue was worked up with ethyl acetate (20 ml) and washed with brine (2x10 ml). The organic part was dried up by adding anhydrous sodium sulfate. The crude product was purified by column chromatography using silica gel(mesh 100-200) and 10% ethylacetate-petroleum ether as eluent to afford pure product. The structure of the product was confirmed by the isolation and characterization of compounds with the help of ^1H and ^{13}C NMR, FT-IR, mass spectrometry, and recording melting pointsof the solid products. Single crystal XRD analyses of **4eb** were also done to establish the structure.

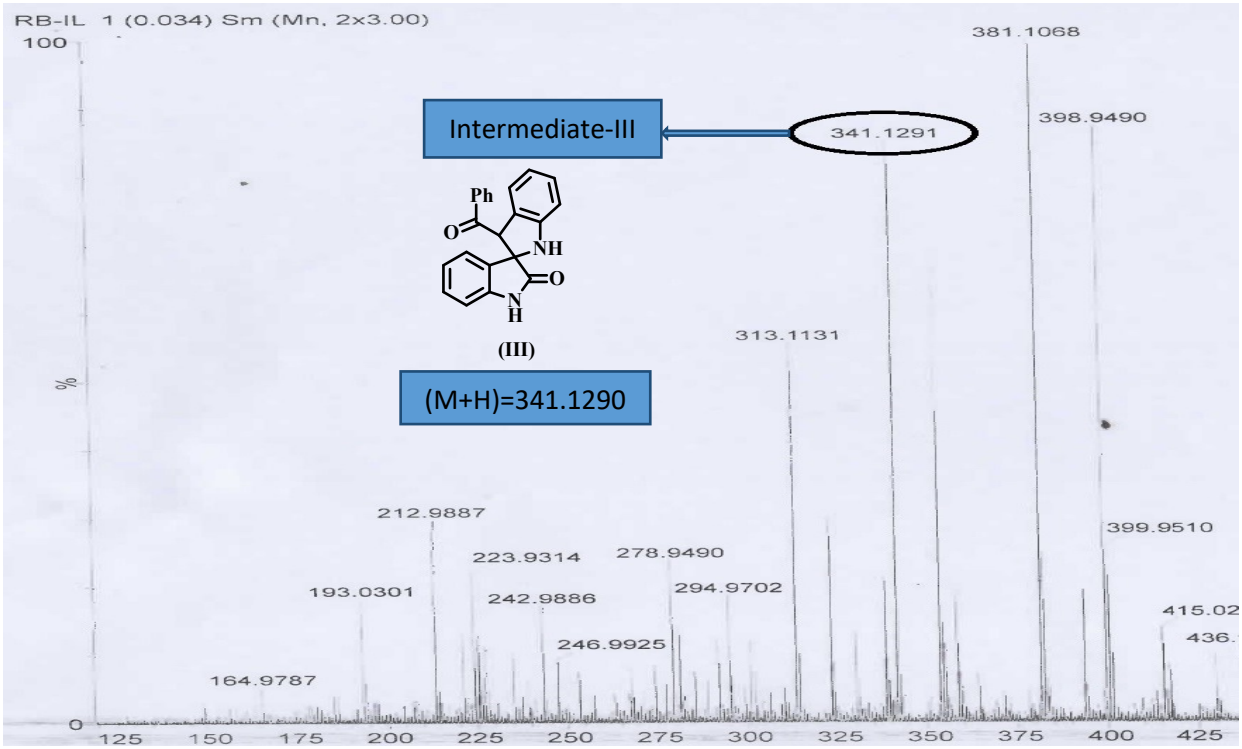
5. HRMS Data Analyses for Intermediate I, II, III and 4aa

To understand the mechanistic pathway we have attempted to collect intermediates from the ongoing reaction mixture. We performed a reaction with **1a (**1.0 mmol) and **2a**(1.0 mmol), 30 mol% of $FeCl_3$ (48.60 mg, 0.30 mmol), 10 mol% of $CuBr_2$ (22.3 mg, 0.10 mmol) in 3 ml dioxane at 120 °C and the reaction was quenched in the middle (after 6 hrs). It was worked up and subjected to a quick filtration column to collect the upper, middle and lower spots (TLC). Those three fractions were subjected to HRMS analysis. Herein, upper spot contains product, middle spot (2^{nd} fraction)may contain intermediate-I and/or intermediate-II and lower spot (3^{rd} fraction) may containrelatively polar intermediate-III. The amounts of 2^{nd} and 3^{rd}collected compounds were insufficient for characterization of the intermediates by NMR.

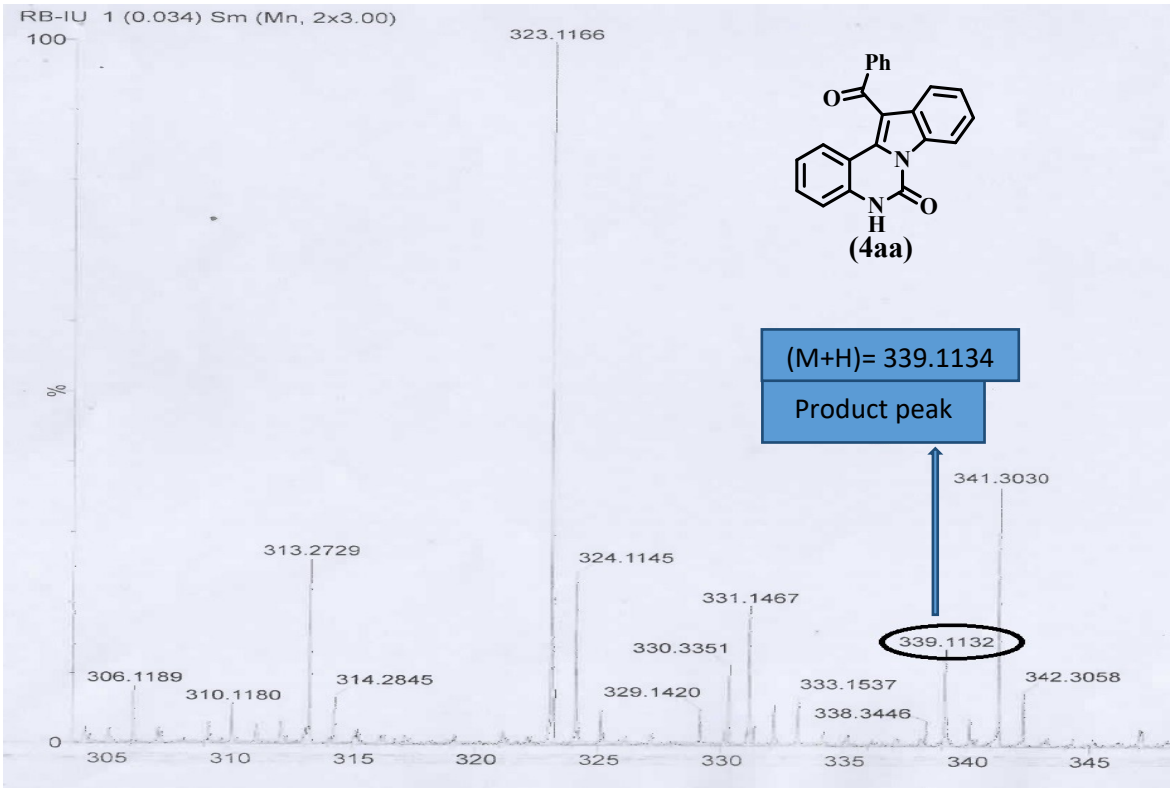
HRMS Data of 2^{nd} Fraction for Intermediate (I) and (II)



HRMS Data of 3rd Fraction for Intermediate (III)



HRMS Data of 1st Fraction for Product (4aa)



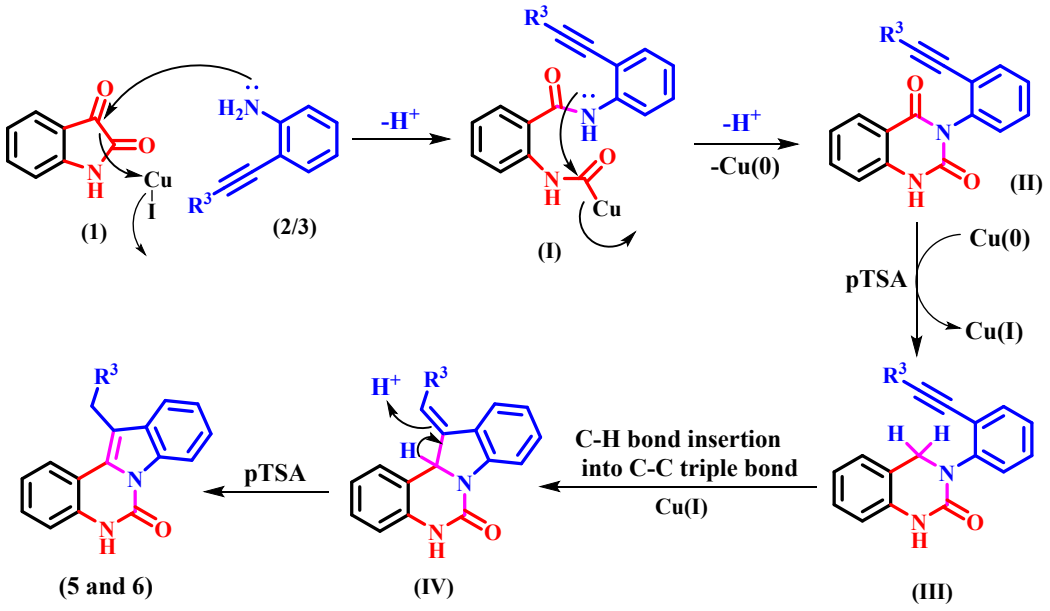
6. General Procedure for the Synthesis of 12-Benzylindolo[1,2-*c*]quinazolin-6(5*H*)-one (5):To a mixture of isatin(**1**, 0.50 mmol) and *o*-alkynylanilie (**3**,0.50 mmol), 20 mol% p-TSA (19.0 mg, 0.10 mmol) and 10 mol% CuI (11.16 mg, .05 mmol) were taken in a oven dried round bottom flask containing a teflon coated magnetic stir bar and 1,4-dioxane (3 ml) was added. The reaction mixture was set to a pre-heated oil bath having temperature 120^{o}C and completion of the reaction was confirmed by monitoring (TLC). The RB flask was picked up from oil and allow to get room temperature.Solvent was removed in rotary evaporator under reduced pressure at room temperature. The residue was worked up with ethyl acetate (20ml) and washed with brine solution (2x10ml). The organic part was dried up by adding anhydrous sodium sulfate and the product was purified by column chromatography using silica gel (mesh 100-200) and 5% ethyl acetate and petroleum ether as an eluent to afford pure product (**5**)**.** The structure of the product was confirmed by the isolation and characterization of compounds with the help of spectroscopic analyses and recording melting points of the solid compounds.

7. General Procedure for the Synthesis of 12-Alkylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6): To a mixture of isatin(**1**, 0.50 mmol) and o-alkynylanilie(**3**,0.50 mmol),20 mol% p-TSA (19.0 mg, 0.10 mmol) and 10 mol% CuI (11.16 mg, .05 mmol, 10 mol%) were taken in a oven dried round bottom flask containing a teflon coated magnetic stir bar and 1,4-dioxane (3 ml) was added. The reaction mixture was set to a pre-heated oil bath having temperature 120^{o}C and completion of the reaction was confirmed by monitoring with TLC. The RB flask was picked up from oil bath and allowed to get room temperature. The solvent was removed in a rotary evaporator under reduced pressure at room temperature. The residue was worked up with ethyl acetate (20ml) and washed with brine solution (2x10ml). The organic part was dried up by adding anhydrous sodium sulfate and the product was purified by column chromatography using silica gel (mesh 100-200) and 5% ethyl acetate in petroleum ether as an eluent to afford pure product (**6**). The structure of the product was confirmed by the isolation and characterization of compounds with the help of spectroscopic analyses and recording melting points of the solid compounds.

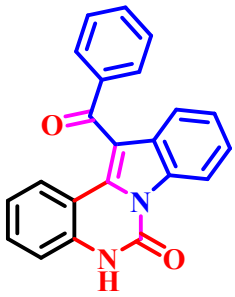
8. Plausible mechanistic pathway to 12-alky/benzylindoloquinazolinone

Here we propose a plausible mechanistic pathway for the p-TSA-Cu(I) combo-catalysis to produce our desired products, **5** and **6**. Firstly, the amino group of 2-ethynyl aniline attacks the ketocarbonyl of isatin, followed by ring opening mediated by Cu-catalysis, leading to the formation of intermediate (**I**). Subsequently, ring closing occurs through further nucleophilic attack of the amido group towards the Cu(I)-activated electrophilic centre, resulting in N-insertion into the isatin ring and the production of an expanded quinazolinone intermediate (**II**). Next, p-TSA activates the amidocarbonyl in close proximity to the triple bond in intermediate (**II**), and it gets reduced through Cu-catalysis to form intermediate -**III**, which further undergoes a cyclization reaction with the triple bond in a proximate position towards the formation of intermediate-**IV**. Finally, to achieve higher stability, the exocyclic double bond is transformed into an endodouble bond in the final products (**5** and **6**) through a simple intramolecular H-shift.

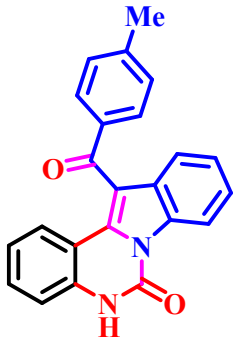
Scheme S2. Plausible mechanism for formation of compound **5** and **6**



9. Spectroscopic Data of Indoloquinazolinones

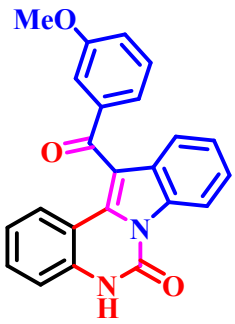


12-Benzoylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4aa) : The compound **(4aa)** was prepared using isatin (0.50 mmol) and 2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound as off white solid (138.8 mg, 0.41 mmol, 82% yield),**M.P.**287-290^{0}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ11.81 (s,1H), 8.71 (d,*J*=8.4 Hz, 1H), 7.93-7.91(m, 2H), 7.71-7.67 (m, 2H), 7.53 (t,*J*=7.6 Hz, 2H), 7.47-7.41 (m, 2H), 7.35-7.27 (m,3H), 7.05-7.01(m, 1H);**^{13}C NMR** (100 MHz, d_6-DMSO): δ198.7, 152.1, 143.2, 140.7, 139.2, 139.0,137.8, 135.8, 134.9, 134.3, 134.0, 130.6, 129.4, 129.3, 127.9, 124.5, 121.2, 121.0, 117.6, 117.4, **FT-IR** (KBr, cm^{-1}):υ_{max}1014, 1049,1141, 1269, 1338, 1385, 1435, 1467, 1509, 1542, 1642, 1728, 2918,3271, 3329;ESI-MS (*m*/*z*) for $C_{22}H_{15}N_2O_2$ $[M+H]^+$: Calculated 339.1134, found 339.1132.

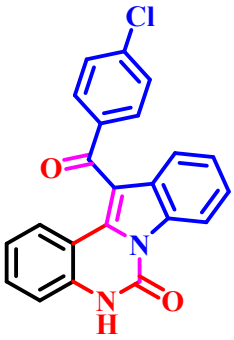


12-(4-Methylbenzoyl)indolo[1,2-c]quinazolin-6(5*H*)-one(4ab) : The compound **(4ab)** was prepared using isatin (0.50 mmol) and 2-(*p*-tolylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound as white solid (146.3 mg, 0.415 mmol, 83% yield), **M.P.**282-284^{o}C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.75 (s, 1H), 8.70 (d, *J*= 8.4Hz, 1H), 7.81 (d, *J* = 7.8 Hz, 2H), 7.67 (d, *J* = 8.1 Hz, 1H),7.46 – 7.25 (m, 7H), 7.06-7.01 (m, 1H), 2.38 (s,3H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 193.5, 147.3, 145.1, 135.9, 135.8, 133.8, 133.0, 130.9, 130.3, 130.1, 129.3, 125.7, 124.6, 124.4, 123.1, 119.8, 116.5, 116.2, 112.9, 112.8, 21.7**FT-IR** (KBr, cm^{-1}):

υ_{max}1009, 1055,1147, 1275,1341, 1388, 1433, 1466, 1511, 1545, 1644, 1730, 2920,3276, 3332;ESI-MS (*m/z*) for $C_{23}H_{17}N_2O_2$ $[M+H]^+$: Calculated 353.1290, found 353.1294.

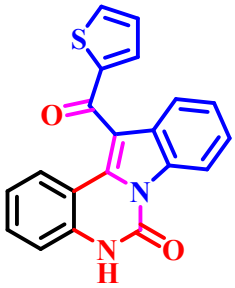


12-(3-Methoxybenzoyl)indolo[1,2-c]quinazolin-6(5*H*)-one (4ac): The compound **(4ac)** was prepared using isatin (0.50 mmol) and 2-[(3-methoxyphenyl)ethynyl]aniline (0.50 mmol) as starting materials. Purification by column chromatography (15% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (154.7 mg, 0.42 mmol, 84% yield), **M.P.**289-290^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.79 (s, 1H), 8.70 (d, *J*= 8.1Hz1H), 7.68 (d, *J*= 8.1Hz1H), 7.49-7.39 (m,5H), 7.37-7.25(m, 4H), 7.04 (t, *J*= 7.8 Hz, 1H), 3.78 (s, 3H); **^{13}C NMR** (100MHz, d_6-DMSO): δ 193.6, 160.1, 147.3, 139.9, 135.9, 134.4, 133.0, 131.0, 130.6, 129.3, 126.0, 124.6, 124.5, 123.0, 123.0, 120.4, 119.8, 116.5, 116.2, 114.1, 112.8, 112.7, 55.8; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1057,1146, 1272, 1344, 1389, 1435, 1464, 1515, 1543, 1648, 1735, 2919,3278, 3335; ESI-MS (*m/z*) for $C_{23}H_{16}N_2NaO_3$ $[M+Na]^+$: Calculated 391.1059, found 391.1056.

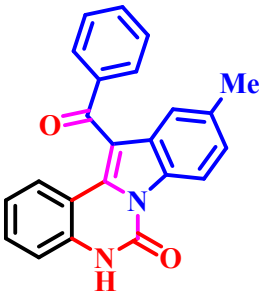


12-(4-Chlorobenzoyl)indolo[1,2-c]quinazolin-6(5*H*)-one (4ad): The compound **(4ad)** was prepared using isatin (0.50 mmol) and 2-[(*p*-chlorophenyl)ethynyl]aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asstraw yellow solid (141.7 mg, 0.38 mmol, 76% yield), **M.P.**309-310^{0}C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.82 (s, 1H), 8.71 (d,*J*=8.4Hz,1H), 7.91 (d,*J*=8.1Hz,2H), 7.67(d, *J* =8.4Hz,1H), 7.59-7.53 (m, 2H), 7.48-7.28 (m, 5H), 7.07-7.02 (m, 1H);**^{13}C NMR**(100 MHz,d_6-DMSO): δ192.6, 147.3, 139.3, 137.2, 136.0, 134.7, 133.1, 132.0, 131.2, 129.7, 129.2, 126.0, 124.7, 124.6, 123.1, 119.8, 116.5, 116.3, 112.7, 112.2, **FT-IR** (KBr,

cm^{-1}): υ_{max}1015, 1058, 1146, 1268, 1341, 1383, 1435, 1462, 1509, 1546, 1637,1717, 2942, 3280, 3351;ESI-MS (*m*/*z*) for $C_{22}H_{14}ClN_2O_2[M+H]^+$: Calculated 373.0744, found 373.0749

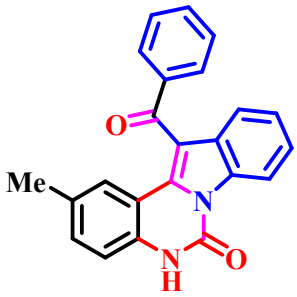


12-(Thiophene-2-carbonyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4ae): The compound **(4ae)** was prepared using isatin (0.50 mmol) and 2-(thiophen-2-ylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff white solid (136.1mg, 0.395 mmol, 79% yield), **M.P.**290-292^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.77 (s, 1H), 8.71-8.68(m,1H), 8.15 (dd, *J* = 5.1 Hz,1.2 Hz, 1H), 7.69-7.62 (m,2H), 7.52-7.31 (m, 5H), 7.18-7.15(m, 1H), 7.11-7.06 (m, 1H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 185.7, 147.3, 144.9, 137.3, 136.8, 135.9, 133.4, 133.0, 130.9, 129.6, 129.1, 125.8, 124.7, 124.6, 123.2, 119.6, 116.4, 116.3, 112.8, 112.6; **FT-IR** (KBr, cm^{-1}): υ_{max}1008, 1065, 1141,1270, 1342, 1385, 1455, 1462, 1522, 1549, 1633,1721, 2935, 3283, 3347;$C_{20}H_{13}N_2O_2S[M+H]^+$: Calculated 345.0698, found 345.0695.

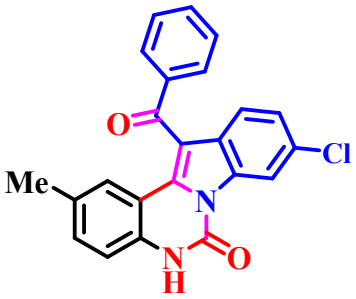


12-Benzoyl-10-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4af): The compound **(4af)** was prepared using isatin (0.50 mmol) and 4-methyl-2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aslight brown solid (146.3mg, 0.415 mmol, 83% yield), **M.P.** 275-277^{o}C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.74 (s, 1H), 8.56 (d,*J*= 8.7 Hz, 1H), 7.91 (d, *J*= 7.5 Hz, 2H), 7.71-7.23 (m,7H), 6.99(t,*J* = 7.8Hz, 2H), 2.34 (s, 3H);**^{13}C NMR** (75 MHz, d_6-DMSO): δ 194.0, 147.2, 138.4, 135.9, 134.4, 134.0, 133.8, 131.3, 130.8, 130.1, 129.5, 126.0, 125.8, 123.0, 119.3, 116.2, 116.1, 112.8, 112.9, 21.6; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1058, 1142, 1273, 1346,

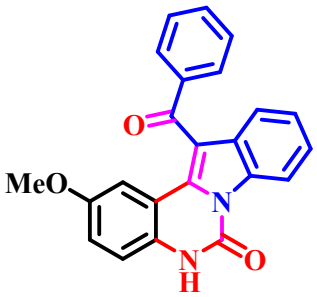
1385, 1437, 1468, 1515, 1549, 1644, 1732, 2922,3275, 3329;ESI-MS (*m*/*z*) for $C_{23}H_{17}N_2O_2$ $[M+H]^+$: Calculated 353.1290, found 353.1292.



12-Benzoyl-2-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4ba): The compound **(4ba)** was prepared using 5-methylisatin(0.50 mmol) and 2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff-white solid (141.1mg, 0.40 mmol, 80% yield), **M.P.** 278-280°C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.72 (br s, 1H), 8.71 (d, *J*= 8.4 Hz, 1H), 7.90 (d, *J*= 7.8Hz, 2H), 7.71-7.66 (m, 1H), 7.54-7.49 (m, 2H), 7.45-7.20 (m, 6H), 2.07 (s, 3H);**^{13}C NMR** (100 MHz, d_6-DMSO): δ 193.8, 147.3, 138.9, 134.7, 134.2, 133.8, 133.1, 132.0, 131.9, 130.0, 129.5, 129.4, 126.2, 124.6, 124.4, 119.9, 116.5, 116.1, 112.6, 112.5, 20.9; **FT-IR** (KBr, cm^{-1}): υ_{max}1009, 1058, 1145, 1271, 1341, 1389, 1440, 1468, 1517, 1549, 1647, 1733, 2932,3277, 3334;ESI-MS (*m*/*z*) for $C_{23}H_{17}N_2O_2$ $[M+H]^+$: Calculated 353.1290, found 353.1294.

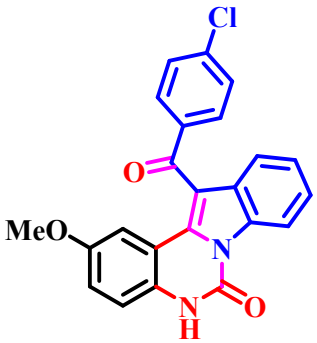


12-Benzoyl-9-chloro-2-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4bg): The compound **(4bg)** was prepared using 5-methylisatin (0.50 mmol) and 5-chloro-2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (145.1 mg, 0.375 mmol, 75% yield), M.P.284-287°C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.80 (br s, 1H), 8.70 (s, 1H), 7.88 (d, *J*= 7.5 Hz, 2H), 7.71-7.66(m, 1H), 7.54 (t,*J*= 7.5 Hz, 2H), 7.37-7.31 (m, 3H), 7.27-7.18 (m, 2H), 2.05 (s, 3H); **^{13}C NMR** (75 MHz, d_6-DMSO): δ 193.2, 147.0, 138.8, 134.2, 133.8, 133.3, 131.9, 130.0, 129.4, 128.7, 128.1, 126.3, 121.2, 116.1, 116.0, 112.4, 112.1, 20.9; **FT-IR** (KBr, cm^{-1}): υ_{max}1011, 1055, 1143, 1270, 1340, 1383, 1434, 1462, 1509, 1551, 1633, 1726, 2938, 3290, 3352; ESI-MS (*m*/*z*) for $C_{23}H_{16}ClN_2O_2$ $[M+H]^+$: Calculated 387.0900, found 387.0897



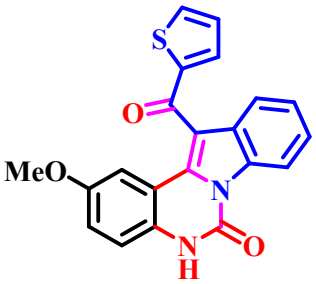
12-Benzoyl-2-methoxyindolo[1,2-c]quinazolin-6(5*H*)-one (4ca): The compound **(4ca)** was prepared using 5-methoxyisatin (0.50 mmol) and 2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (145.6 mg, 0.395 mmol, 79% yield), **M.P.** 285-287°C;

^{1}H NMR (300 MHz, d_6-DMSO): δ 11.69 (s, 1H), 8.73 (d,*J*= 8.1 Hz, 1H), 7.93 (d, *J*= 7.5Hz, 2H), 7.68 (t,*J*= 7.2 Hz,1H), 7.54-7.25(m,6H), 7.10-7.06 (m, 2H), 3.38 (s, 3H);**^{13}C NMR** (75 MHz, d_6-DMSO): δ 193.5, 154.7, 147.1, 138.8, 134.4, 134.3, 133.2, 130.0, 129.9, 129.6, 129.4, 124.7, 124.5, 119.9, 119.4, 117.5, 116.5, 113.2, 112.6, 109.1, 55.5; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1059,1149, 1277, 1342, 1388, 1438, 1465, 1513, 1545, 1641, 1728, 2928,3276, 3343; ESI-MS (*m*/*z*) for $C_{23}H_{17}N_2O_3[M+H]^+$: Calculated 369.1239, found 369.1244.



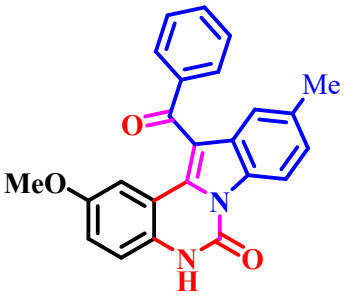
12-(4-Chlorobenzoyl)-2-methoxyindolo[1,2-*c*]quinazolin-6(5*H*)-one (4cd): The compound **(4cd)** was prepared using 5-methoxyisatin (0.50 mmol) and 2-[(*p*-chlorophenyl)ethynyl]aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aswhitesolid (149.1 mg, 0.37 mmol, 74% yield),**M.P.** 285-289°C;**^{1}H NMR** (400 MHz, $CDCl_3$): δ 11.73 (s, 1H), 8.73 (d,*J*= 8.4 Hz,1H), 7.93 (d,*J*= 8Hz,2H), 7.56 (d,*J*= 8Hz,2H), 7.46-7.33 (m, 3H), 7.27 (d, *J*= 9.2Hz,1H), 7.11 (d,*J*= 7.2 Hz,2H), 3.44 (s, 3H); **^{13}C NMR** (75 MHz, $CDCl_3$): δ192.2, 154.7, 147.1, 139.2, 137.5, 134.9, 133.2, 132.0, 130.1, 129.7, 129.3, 124.7, 124.6, 119.9, 119.6, 117.6, 116.5, 113.1, 112.1, 109.1, 55.5;**FT-IR** (KBr, cm^{-1}): υ_{max}1015, 1048,1142, 1267, 1338, 1384, 1436, 1470, 1512, 1542, 1645,

1731, 2934,3269, 3340;ESI-MS (*m*/*z*) for $C_{23}H_{16}ClN_2O_3$ $[M+H]^+$: Calculated 403.0849, found 403.0844.



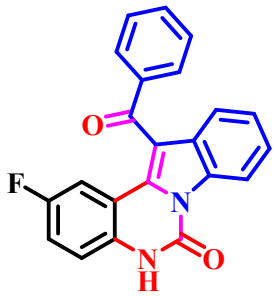
2-Methoxy-12-(thiophene-2-carbonyl)indolo[1,2-c]quinazolin-6(5*H*)-one (4ce): The compound **(4ce)** was prepared using 5-methoxyisatin (0.50 mmol) and 2-(thiophen-2-ylethynyl)aniline(0.50 mmol)

as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asgreenish yellow solid (142.4 mg, 0.38 mmol, 76% yield), **M.P.** 287-289^{0}C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.67 (s, 1H), 8.73-8.69 (m,1H), 8.15(dd, *J* = 5.1 Hz, 1.2 Hz,1H), 7.66 (dd, *J*=3.9Hz, 1.2Hz,1H), 7.60-7.57 (m,1H), 7.48-7.37 (m, 2H), 7.27 (dd, *J* = 5.4Hz, 4.2Hz, 1H), 7.19-7.16 (m,1H), 7.12-7.08 (m, 2H), 3.47 (s, 3H);**^{13}C NMR** (100 MHz, d_6-DMSO): δ 185.4, 154.7, 147.1, 145.0, 137.2, 136.8, 133.6, 133.1, 129.9, 129.7, 129.1, 124.7, 124.6, 119.6, 119.2, 117.7, 116.5, 113.1, 112.5, 108.8, 55.4; **FT-IR** (KBr, cm^{-1}): υ_{max}1016, 1067, 1141, 1272, 1346, 1385, 1459, 1462, 1523, 1547, 1635, 1727, 2935, 3281, 3338; ESI-MS (*m*/*z*) for $C_{21}H_{14}N_2NaO_3S$ $[M+Na]^+$: Calculated 397.0623, found 397.0619.

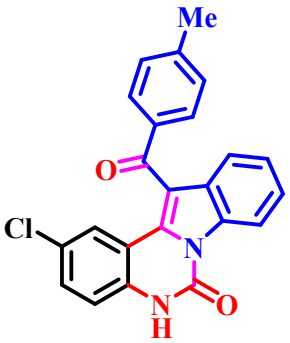


12-Benzoyl-2-methoxy-10-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4cf): The compound **(4cf)** was prepared using 5-methoxyisatin (0.50 mmol) and 4-methyl-2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff-white solid (153.0 mg, 0.40 mmol, 80% yield), **M.P.** 270-272 ^{0}C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.64 (s, 1H), 8.58 (d,*J*= 8.4 Hz, 1H), 7.92 (d, *J*= 7.8Hz, 2H), 7.69(d,*J*= 7.2 Hz,1H), 7.54-7.49 (m, 2H), 7.27-7.22 (m, 3H), 7.06-6.97 (m, 2H) 3.34 (s, 3H), 3.37 (s, 3H);**^{13}C NMR** (75 MHz, d_6-DMSO): δ 195.5, 154.7, 147.0, 138.7, 134.3, 133.8, 131.5, 130.0, 129.9, 129.7, 129.6, 126.0, 119.4, 119.3, 117.5, 116.1, 113.2,

112.2, 108.9, 55.4, 21.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1014, 1067, 1142, 1271, 1348, 1385, 1464, 1523, 1551, 1640, 1736, 2931, 3276, 3330; ESI-MS (*m*/*z*) for $C_{24}H_{19}N_2O_3[M+H]^+$: Calculated 383.1396, found 383.1393.

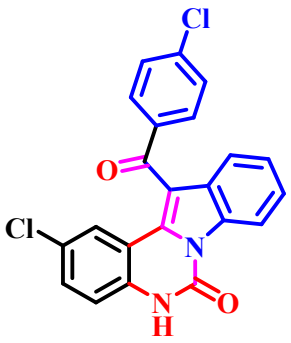


12-Benzoyl-2-fluoroindolo[1,2-c]quinazolin-6(5*H*)-one (4da): The compound **(4da)** was prepared using 5-fluoroisatin (0.50 mmol) and 2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff white solid (151.5 mg, 0.425 mmol, 85% yield), **M.P.** 277-279°C;**^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.86 (s, 1H), 8.71 (d,*J*= 8.1Hz, 1H), 7.89 (d, *J*= 7.8Hz, 2H), 7.71(t,*J*= 7.5 Hz,1H), 7.56-7.20 (m, 8H);**^{13}C NMR** (100 MHz, d_6-DMSO): δ 193.8, 157.5 (C-F, $^1J_{C\text{-}F}$= 237 Hz), 147.0, 138.7, 134.5, 133.8, 133.1, 132.7, 130.1, 129.5, 129.2, 124.9, 124.8, 120.2, 118.8 (C-F, $^2J_{C\text{-}F}$= 24 Hz), 118.1 (C-F, $^3J_{C\text{-}F}$= 8 Hz), 116.6, 113.9 (C-F, $^3J_{C\text{-}F}$= 9 Hz), 113.5, 111.4 (C-F, $^2J_{C\text{-}F}$= 26 Hz); **FT-IR** (KBr, cm^{-1}): υ_{max}1010, 1054, 1143, 1268, 1340, 1382, 1435, 1458, 1507, 1547, 1628, 1718, 2940, 3286, 3351; ESI-MS (*m*/*z*) for $C_{22}H_{14}FN_2O_2$ $[M+H]^+$: Calculated 357.1039found 357.1036.

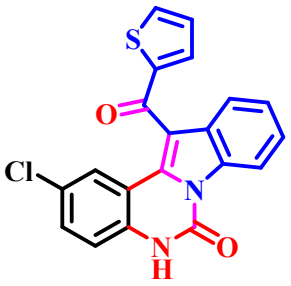


2-Chloro-12-(4-methylbenzoyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4eb): The compound **(4eb)** was prepared using 5-chloroisatin (0.50 mmol) and 2-(*p*-tolylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (164.5 mg, 0.425 mmol, 85% yield), **M.P.**280-282°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.90 (s, 1H), 8.70 (d, *J*= 8.4 Hz, 1H), 7.81-7.74 (m,3H),

7.53-7.41 (m, 2H), 7.36-7.30 (m,4H), 7.24 (d, *J*= 8.1 Hz, 1H), 2.40 (s, 3H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 193.3, 147.0, 145.1, 136.1, 134.8, 133.0, 132.9, 130.7, 130.3, 130.1, 129.2, 126.7, 124.9, 124.8, 124.7, 120.2, 118.0, 116.5, 114.4, 113.7, 21.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1060, 1141, 1274, 1341, 1381, 1434, 1465, 1512, 1555, 1632, 1725, 2936, 3288, 3348; ESI-MS (*m*/*z*) for $C_{23}H_{16}ClN_2O_2$ $[M+H]^+$: Calculated 387.0900, found 387.0898.

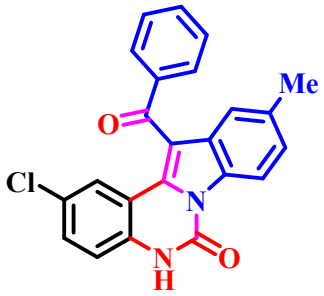


2-Chloro-12-(4-chlorobenzoyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4ed): The compound **(4ed)** was prepared using 5-chloroisatin (0.50 mmol) and 2-[(*p*-chlorophenyl)ethynyl]aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (152.8 mg, 0.375 mmol, 75% yield), **M.P.**305-307^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.96 (s, 1H), 8.70 (d, *J*= 8.1 Hz, 1H), 7.91-7.89 (m, 2H), 7.77 (d,*J*= 2.4Hz,1H), 7.63-7.59 (m, 2H), 7.54-7.51 (m, 1H), 7.47-7.42 (m,1H), 7.36, 7.25 (m, 3H); **^{13}C NMR** (75 MHz, , d_6-DMSO): δ 192.4, 147.0, 139.3, 137.4, 134.9, 133.9, 133.1, 132.0, 131.0, 129.6, 129.0, 126.7, 125.2, 124.9, 124.8, 120.2, 118.0, 116.5, 114.2, 113.1;**FT-IR** (KBr,cm^{-1}): υ_{max}1011, 1052, 1140, 1263, 1341, 1378, 1432, 1460, 1506, 1536, 1625, 1714, 2942, 3287, 3356; ESI-MS (*m*/*z*) for $C_{22}H_{13}Cl_2N_2O_2$ $[M+H]^+$: Calculated 407.0354, found 407.0359.

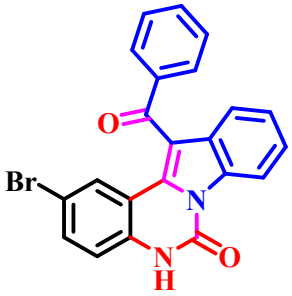


2-Chloro-12-(thiophene-2-carbonyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4ee): The compound **(4ee)** was prepared using 5-chloroisatin (0.50 mmol) and 2-(thiophen-2-ylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff white solid (155.4 mg, 0.41 mmol, 82% yield), **M.P.**

295-297°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.90 (s, 1H), 8.71-8.68 (m,1H), 8.20 (dd, *J* = 5.1 Hz, 1.2 Hz,1H), 7.67-7.64 (m, 2H), 7.54-7.31(m, 5H*)*), 7.21-7.19 (m, 1H); **^{13}C NMR** (100 MHz, d_6-DMSO): δ 185.4, 147.0, 144.7, 137.5, 137.1, 134.8, 133.0, 132.4, 130.7, 129.7, 128.9, 126.8, 125.0, 124.9, 124.8, 119.9, 118.1, 116.5, 114.3, 113.4; **FT-IR** (KBr, cm^{-1}): υ_{max}1007, 1063, 1140, 1269, 1342, 1383, 1456, 1461, 1520, 1550, 1630, 1719, 2937, 3282, 3348;ESI-MS (*m*/*z*) for $C_{20}H_{12}ClN_2O_2S$ $[M+H]^+$: Calculated 379.0308, found 379.0310.

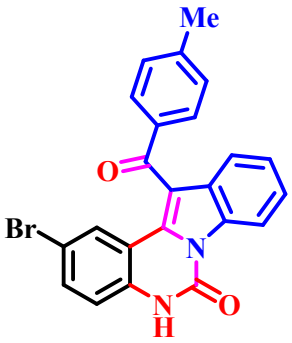


12-Benzoyl-2-chloro-10-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (4ef): The compound **(4ef)** was prepared using 5-chloroisatin (0.50 mmol) and 4-methyl-2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff white solid (164.5 mg, 0.425 mmol, 85% yield),**M.P.**290-292°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.87 (s, 1H), 8.55 (d,*J*= 8.7 Hz, 1H), 7.89 (d, *J*= 7.8 Hz, 2H), 7.74-7.64(m, 2H), 7.56-7.45(m, 3H), 7.31-7.24 (m, 2H), 7.06 (s, 1H) 2.32 (s, 3H); **^{13}C NMR** (100 MHz, d_6-DMSO): δ 193.8, 147.0, 138.5, 134.8, 134.5, 134.0, 133.2, 131.4, 130.7, 130.1, 129.6, 129.4, 126.7, 126.5, 125.0, 119.7, 118.0, 116.2, 114.4, 113.2, 21.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1011, 1061, 1144, 1275, 1341, 1383, 1434, 1466, 1510, 1557, 1631, 1726, 2935, 3289, 3346; ESI-MS (*m*/*z*) for $C_{23}H_{16}ClN_2O_2$ $[M+H]^+$: Calculated 387.0900, found 387.0896.

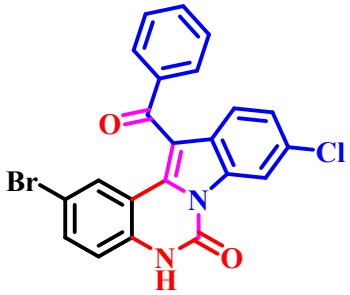


12-Benzoyl-2-bromoindolo[1,2-*c*]quinazolin-6(5*H*)-one (4fa): The compound **(4fa)** was prepared using 5-bromoisatin (0.50 mmol) and 2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound as brownish solid (173.2 mg, 0.415 mmol, 83% yield), **M.P.**300-304°C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.92(s, 1H), 8.69(d, *J*= 8.4 Hz, 1H), 7.91-7.87

(m,3H), 7.73-7.69(m, 1H), 7.61-7.52 (m, 3H), 7.43(t, *J*=7.6 Hz, 1H), 7.33-7.30(m,1H), 7.27-7.24(m, 2H); **^{13}C NMR** (100 MHz, d_6-DMSO): δ 193.7, 147.0, 138.7, 135.2, 134.4, 133.5, 133.4, 133.1, 130.1, 129.6, 129.2, 128.1, 124.9, 124.8, 120.2, 118.3, 116.5, 114.8, 114.5, 113.5; **FT-IR** (KBr, cm^{-1}): υ_{max}1007, 1056, 1144, 1271, 1340, 1387, 1440, 1466, 1516, 1549, 1645, 1730, 2929,3278, 3337; ESI-MS (*m*/*z*) for $C_{22}H_{14}BrN_2O_2$ $[M+H]^+$: Calculated 417.0239, found 417.0237.

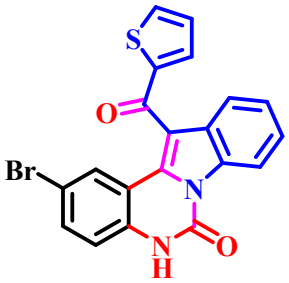


2-Bromo-12-(4-methylbenzoyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4fb): The compound **(4fb)** was prepared using 5-bromoisatin (0.50 mmol) and 2-(*p*-tolylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff white solid (183.3 mg, 0.425 mmol, 85% yield), **M.P.** 290-295^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.90 (s, 1H), 8.70 (d, *J*=8.1Hz, 1H), 7.87-7.79 (m, 3H), 7.6 (dd, *J*=8.7Hz, 2.4Hz,1H), 7.47-7.25 (m, 6H), 2.40 (s, 3H); **^{13}C NMR**(75MHz, d_6-DMSO): δ 193.3, 147.0, 145.1, 136.1, 135.1, 133.4, 133.0, 132.9, 130.2, 130.1, 129.2, 127.9, 124.8, 124.7, 120.2, 118.2, 116.5, 114.8, 114.5, 113.7, 21.8; **FT-IR** (KBr, cm^{-1}): υ_{max}1008, 1057, 1144, 1273, 1342, 1387, 1443, 1467, 1516, 1550, 1646, 1731, 2933,3279, 3335;ESI-MS (*m*/*z*) for $C_{23}H_{16}BrN_2O_2$ $[M+H]^+$: Calculated 431.0395 found 431.0399.



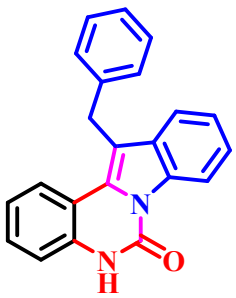
12-Benzoyl-2-bromo-9-chloroindolo[1,2-*c*]quinazolin-6(5*H*)-one (4fg): The compound **(4fg)** was prepared using 5-bromoisatin (0.50 mmol) and 4-chloro-2-(phenylethynyl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound as light brownsolid (169.5 mg, 0.375 mmol, 75% yield), **M.P.** 298-

300°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 12.02(s, 1H), 8.69 (s, 1H), 7.90-7.84 (m,3H), 7.74-7.69(m, 1H), 7.64-7.52 (m, 3H), 7.38 (d, *J*= 8.7 Hz, 1H), 7.25 (d, *J*= 9.0 Hz, 2H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 193.1, 146.8, 138.6, 135.2, 134.5, 134.3, 133.8, 133.3, 130.1, 129.6, 129.2, 128.3, 127.9, 125.0, 121.6, 118.3, 116.0, 114.6, 113.1; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1053, 1142, 1264, 1344, 1376, 1434, 1461, 1506, 1537, 1627, 1716, 2939, 3285, 3354; ESI-MS (*m*/*z*) for $C_{22}H_{13}BrClN_2O_2$ $[M+H]^+$: Calculated 450.9849, found 450.9854.

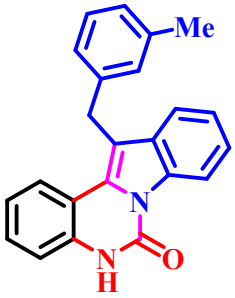


2-Bromo-12-(thiophene-2-carbonyl)indolo[1,2-*c*]quinazolin-6(5*H*)-one (4fe): The compound **(4fe)** was prepared using 5-bromoisatin (0.50 mmol) and 2-(thiophen-2-ylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound asoff whitesolid (169.3 mg, 0.40 mmol, 80% yield), **M.P.** 285-287°C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ11.90(s, 1H), 8.69 (d, *J*=8 Hz,1H), 8.20 (d, *J*= 4.8 Hz, 1H), 7.80 (d, *J*= 2.4 Hz, 1H), 7.65-7.59 (m, 2H), 7.54-7.37 (m, 3H), 7.25 (d, *J*= 8.8 Hz, 1H), 7.20 (t, *J*= 4.4 Hz, 1H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ185.3, 147.0, 144.7, 137.5, 137.0, 135.1, 133.4, 133.0, 132.3, 129.7, 128.9, 128.0, 125.0, 124.8, 119.9, 118.4, 116.5, 114.7, 114.5, 113.3; **FT-IR** (KBr, cm^{-1}): υ_{max}1007, 1053, 1142, 1270, 1340, 1384, 1442, 1463, 1515, 1548, 1643, 17325, 2926,3275, 3339;ESI-MS (*m*/*z*) for $C_{20}H_{12}BrN_2O_2S$ $[M+H]^+$: Calculated 422.9803, found 422.9805.

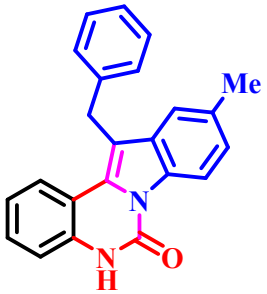
Spectroscopic data of 12-benzylindolo[1,2-c]quinazolin-6(5*H*)-ones



12-Benzylindolo[1,2-*c*]quinazolin-6(5*H*)-one (5aa): The compound **(5aa)** was prepared using isatin (0.50 mmol) and 2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (129.8 mg, 0.40 mmol, 80% yield),**M.P.**260-262^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.37 (s, 1H), 8.63 (d, *J*= 7.5 Hz, 1H), 7.97 (d, *J*= 8.1 Hz, 1H), 7.76 (d, *J*= 7.2 Hz, 1H), 7.43-7.11 (m, 10H), 4.60 (s, 2H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 147.6, 139.8, 135.2, 132.9, 131.2, 129.8, 129.3, 129.0, 128.3, 126.6, 124.4, 124.1, 123.8, 123.3, 118.8, 116.1, 115.9, 114.8, 112.1, 46.1; **FT-IR** (KBr, cm^{-1}): υ_{max}1015, 1051,1143, 1269, 1340, 1385, 1439, 1467, 1509, 1542, 1641, 2936,3272, 3326;ESI-MS (*m*/*z*) for $C_{22}H_{17}N_2O$ $[M+H]^+$: Calculated 325.1341, found 325.1337.

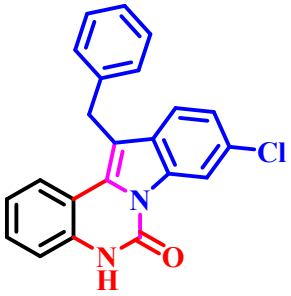


12-(3-Methylbenzyl)indolo[1,2-c]quinazolin-6(5*H*)-one (5ah): The compound **(5ah)** was prepared using isatin (0.50 mmol) and 2-(*m*-tolylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (140.5 mg, 0.415 mmol, 83% yield), **M.P.**248-250 ^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.36 (s, 1H), 8.63 (d, *J*= 7.5 Hz, 1H), 7.96 (d, *J*= 7.8 Hz, 1H), 7.74 (d, *J*= 6.9 Hz, 1H), 7.42-7.26 (m, 4H), 7.16-6.95 (m, 5H), 4.55 (s, 2H), 2.19 (s, 3H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 147.6, 139.7, 138.0, 135.2, 133.0, 131.3, 129.8, 129.3, 128.9, 127.3, 125.4, 124.5, 124.1, 123.8, 123.3, 118.8, 116.1, 115.9, 114.8, 112.2, 30.0, 21.5; **FT-IR** (KBr, cm^{-1}): υ_{max}1010, 1053,1143, 1275, 1342, 1386, 1438, 1464, 1511, 1545, 1635, 2938,3273, 3345; ESI-MS (*m*/*z*) for $C_{23}H_{19}N_2O$ $[M+H]^+$: Calculated 339.1497, found 339.1500.

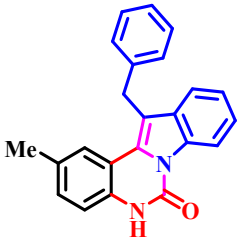


12-Benzyl-10-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (5af): The compound **(5af)** was prepared using isatin (0.50 mmol) and 4-methyl-2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded

the title compound as white solid (140.5 mg, 0.415 mmol, 83% yield), **M.P.**270-272°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.33 (s, 1H), 8.50 (d, *J*= 8.4 Hz, 1H), 7.92 (d, *J*= 8.1 Hz, 1H), 7.54 (s, 1H), 7.39-7.09 (m, 9H), 4.57 (s, 2H), 2.44 (s, 3H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 147.5, 139.8, 135.1, 132.9, 131.5, 131.2, 129.9, 129.2, 129.0, 128.3, 126.6, 125.6, 124.4, 123.3, 118.3, 115.8, 114.9, 111.8, 30.0, 21.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1012, 1059, 1142, 1275, 1345, 1386, 1437, 1469,1518, 1550, 1647, 2935,3277, 3332;ESI-MS (*m*/*z*) for $C_{23}H_{19}N_2O$ $[M+H]^+$: Calculated 339.1497, found 339.1495.

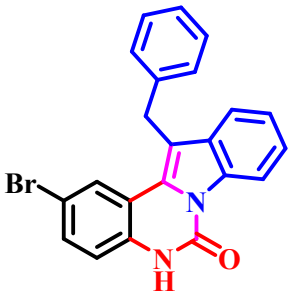


12-Benzyl-9-chloroindolo[1,2-*c*]quinazolin-6(5*H*)-one (5ag): The compound **(5ag)** was prepared using isatin (0.50 mmol) and 4-chloro-2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (136.4 mg, 0.38 mmol, 76% yield), **M.P.** 286-287°C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.47 (br s, 1H), 8.62 (s, 1H), 7.95 (d, *J*= 8.1 Hz, 1H), 7.76 (d, *J*= 8.4 Hz, 1H), 7.42-7.37 (m, 2H), 7.28-7.11 (m, 7H), 4.58 (s, 2H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.4,139.6, 135.1, 133.1, 130.7, 130.0, 129.6, 129.0, 128.4, 128.3, 126.7, 124.5, 124.0, 123.5, 120.3, 116.0, 115.7, 114.6, 112.0, 46.2; **FT-IR** (KBr, cm^{-1}): υ_{max}1011, 1051, 1146, 1265, 1341, 1380, 1435, 1460, 1509, 1543, 1637, 2941, 3284, 3355; ESI-MS (*m*/*z*) for $C_{22}H_{16}ClN_2O$ $[M+H]^+$: Calculated 359.0951, found 359.0949.

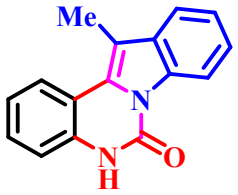


12-Benzyl-2-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (5ba): The compound **(5ba)** was prepared using 5-methylisatin (0.50 mmol) and 2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (130.3 mg, 0.385 mmol, 77% yield), **M.P.**274-278 °C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.27 (s, 1H), 8.65- 8.62 (m, 1H), 7.78 (d, *J*= 5.7 Hz, 2H), 7.39-7.36 (m, 2H), 7.25-7.14 (m, 7H), 4.59 (s, 2H), 2.25 (s, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.6,

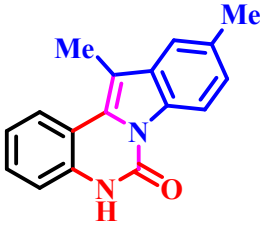
140.1, 132.9, 132.9, 132.2, 131.4, 130.1, 129.7, 129.0, 128.5, 126.6, 124.6, 124.0, 123.8, 118.8, 116.1, 115.7, 114.6, 112.3, 30.2, 21.1; **FT-IR** (KBr, cm^{-1}): υ_{max}1008, 1056,1147, 1278, 1341, 1389, 1431, 1463, 1511, 1544, 1649, 2933,3276, 3328;ESI-MS (*m*/*z*) for $C_{23}H_{19}N_2O$ $[M+H]^+$: Calculated 339.1497, found 339.1494.



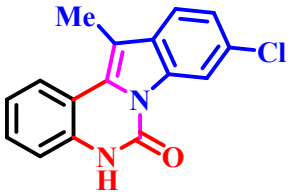
12-Benzyl-2-bromolindolo[1,2-*c*]quinazolin-6(5*H*)-one (5fa): The compound **(5fa)** was prepared using 5-bromoisatin (0.50 mmol) and 2-(phenylethynyl)aniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (161.4 mg, 0.40 mmol, 80% yield), **M.P.** 278-280°C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.49 (s, 1H), 8.64- 8.62 (m, 1H), 7.96 (d, *J*= 2.4 Hz, 1H), 7.85-7.83 (m, 1H), 7.53 (dd,*J* = 8.8Hz, 2.0 Hz, 1H), 7.46-7.39 (m, 2H), 7.29-7.18 (m, 6H), 4.58 (s, 2H); **^{13}C NMR** (75 MHz,d_6-DMSO): δ 147.3, 139.5, 134.4, 133.0, 131.7, 131.2, 129.1, 128.4, 128.3, 126.7, 126.6, 124.6, 124.1, 119.0, 117.8, 116.7, 116.1, 114.9, 113.4, 30.0; **FT-IR** (KBr, cm^{-1}): υ_{max}1006, 1054, 1140, 1273, 1340, 1385, 1440, 1466, 1526, 1549, 1640, 2937,3279, 3342;ESI-MS (*m*/*z*) for $C_{22}H_{16}BrN_2O$ $[M+H]^+$: Calculated 403.0446, found 403.0450.



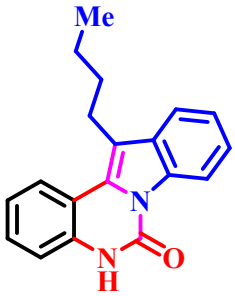
12-Methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6aa): The compound (**6aa**) was prepared using isatin (0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as off white solid (96.9 mg, 0.39 mmol, 78% yield), **M.P.** 295-298°C; **^{1}HNMR** (400 MHz, d_6-DMSO): δ 11.26 (s, 1H), 8.57-8.55 (m, 1H), 8.12-8.10 (m, 1H), 7.79-7.77 (m,1H), 7.43-7.35 (m, 3H), 7.29-7.24 (m, 2H), 2.66 (s, 3H); **^{13}C NMR** (75 MHz, , d_6-DMSO): δ 147.6, 135.0, 132.7, 131.3, 128.9, 124.6, 123.8, 123.5, 123.3, 118.7, 115.9, 115.7, 115.5, 109.3, 10.8; **FT-IR** (KBr, cm^{-1}): υ_{max} 1064, 1131, 1247, 1330, 1377, 1410, 1490, 1595, 1645, 2927, 3085, 3215, 3422; ESI-MS (*m*/*z*) for $C_{16}H_{12}N_2NaO$ $[M+Na]^+$: Calculated 271.0847, found 271.0851.



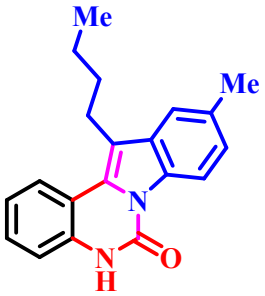
10,12-Dimethylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ab): The compound (**6ab**) was prepared using isatin (0.50 mmol), 2-ethynyl-4-methylaniline (0.50 mmol) and as a startingmaterials. Purification by column chromatography (5% ethyl acetate in petroleumether) afforded the title compound as white solid (105.0 mg, 0.40 mmol, 80% yield), **M.P.** 284-285 ^{o}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.20 (s, 1H), 8.41(d,*J*= 8.4 Hz, 1H), 8.08 (d,*J*= 7.6 Hz, 1H), 7.53 (s, 1H), 7.39 (t,*J*= 7.6 Hz, 1H), 7.26-7.22 (m, 2H), 7.18-7.16 (m, 1H), 2.62 (s, 3H), 2.46 (s, 3H); **^{13}C NMR** (100 MHz, , d_6-DMSO): δ 147.5, 134.9, 132.5, 131.5, 131.0, 128.9, 128.8, 125.3, 124.5, 123.3, 118.3, 115.7, 115.6, 115.5, 109.0, 21.7, 10.8; **FT-IR** (KBr, cm^{-1}): υ_{max}1067, 1136, 1242, 1334, 1370, 1414, 1492, 1597, 1646, 2924, 3081, 3212, 3424; ESI-MS (*m*/*z*) for $C_{17}H_{15}N_2O$ $[M+H]^+$: Calculated 263.1184, found 263.1185.



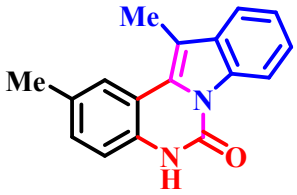
12-Methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ac): The compound **(6ac)** was prepared using isatin (0.50 mmol) and 5-chloro-2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (106.1 mg, 0.375 mmol, 75% yield), **M.P.** 298-300^{o}C; **^{1}HNMR** (400 MHz, d_6-DMSO): δ 11.35 (s, 1H), 8.53 (s, 1H), 8.07 (d, *J*= 8 Hz, 1H), 7.77 (d,*J*= 8.4 Hz, 1H), 7.43-7.37 (m, 2H), 7.28-7.24 (m, 2H), 2.63 (s, 3H); **^{13}C NMR** (100 MHz, , d_6-DMSO): δ 147.4, 134.9, 132.8, 130.0, 129.8, 129.2, 128.1, 124.6, 123.7, 123.5, 120.1, 115.8, 115.4, 115.3, 109.1, 10.8; **FT-IR** (KBr, cm^{-1}): $_{max}$1065, 1134, 1239, 1333, 1370, 1412, 1492, 1596, 1642, 2925, 3083, 3213, 3423; ESI-MS (*m*/*z*) for $C_{16}H_{11}ClN_2NaO$ $[M+Na]^+$: Calculated 305.0458, found 305.0455.



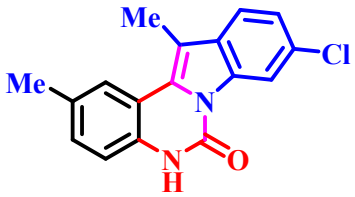
12-Butylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ad): The compound **(6ad)** was prepared using isatin (0.50 mmol) and 2-(pent-1-yn-1-yl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (68.18 mg, 0.235 mmol, 47% yield), **M.P.** 200-202°C; **^{1}HNMR** (400 MHz, d_6-DMSO): δ 11.31-11.28 (m, 1H), 8.60-8.57 (m, 1H), 8.05-8.03 (m, 1H), 7.89-7.74 (m, 1H), 7.45-7.26 (m, 5H), 3.20-3.15 (m, 2H), 1.70-1.64 (m, 2H), 1.51-1.45 (m, 2H), 0.97-0.92 (m, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.7, 135.0, 132.8, 131.0, 129.0, 128.5, 124.2, 123.9, 123.6, 123.5, 118.7, 116.0, 115.9, 115.1, 114.8, 31.8, 24.3, 22.7, 14.4; **FT-IR** (KBr, cm^{-1}): υ_{max}1063, 1133, 1247, 1331,1375, 1412, 1491, 1594, 1644, 2926, 3085, 3214, 3420; ESI-MS (*m*/*z*) for $C_{19}H_{19}N_2O$ $[M+H]^+$: Calculated 291.1497, found 291.1501.



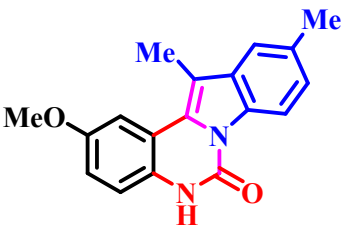
12-Butyl-10-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ae): The compound **(6ae)** was prepared using isatin (0.50 mmol) and 2-(pent-1-yn-1-yl)aniline(0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (74.5 mg, 0.245 mmol, 49% yield), **M.P.** 215-217°C; **^{1}HNMR** (300 MHz, d_6-DMSO): δ 11.23 (s, 1H), 8.44 (d, *J*= 8.4 Hz, 1H), 8.02 (d, *J*= 8.1 Hz, 1H), 7.54 (s, 1H), 7.42-7.37 (m, 1H), 7.28-7.17 (m, 3H), 3.12 (t, *J*= 7.5 Hz, 2H) 2.47 (s, 3H), 1.68-1.63 (m, 2H), 1.48 (t, *J*= 7.5 Hz, 2H), 0.94 (t, *J*= 7.2 Hz, 3H); **^{13}C NMR** (75 MHz, , d_6-DMSO): δ 147.6, 135.0, 132.6, 131.2, 131.1, 128.9, 128.5, 125.4, 124.1, 123.4, 118.2, 115.9, 115.7, 115.2, 114.5, 31.8, 24.3, 22.7, 21.8, 14.4; **FT-IR** (KBr, cm^{-1}): υ_{max}1065, 1134, 1247, 1331, 1374, 1411, 1491, 1592, 1646, 2927, 3083, 3213, 3422;ESI-MS (*m*/*z*) for $C_{20}H_{21}N_2O$ $[M+H]^+$: Calculated 305.1654, found 305.1650.



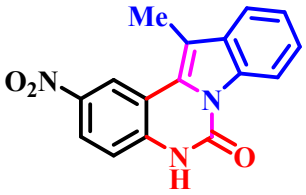
2,12-Dimethylindolo[1,2-c]quinazolin-6(5*H*)-one (6ba): The compound **(6ba)** was prepared using 5-methylisatin (0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (98.5 mg, 0.375 mmol, 75% yield), **M.P.** 280-282 ^{o}C; **^{1}HNMR** (300 MHz, d_6-DMSO): δ 11.14 (s, 1H), 8.58-8.55 (m, 1H), 7.89 (s, 1H), 7.79-7.76 (m,1H), 7.40-7.33(m, 2H), 7.23-7.15 (m, 2H), 2.67 (s, 3H), 2.40 (s, 3H); **^{13}C NMR** (100 MHz, d_6-DMSO): δ 147.6, 132.7, 132.3, 131.3, 129.8, 128.9, 124.5, 123.7, 123.4, 118.6, 115.9, 115.6, 115.4, 109.1, 21.3, 10.9; **FT-IR** (KBr, cm^{-1}): υ_{max}1066, 1135, 1243, 1333, 1372, 1415, 1491, 1597, 1645, 2925, 3081, 3210, 3425; ESI-MS (*m*/*z*) for $C_{17}H_{15}N_2O$ $[M+H]^+$: Calculated 263.1184, found 263.1188.



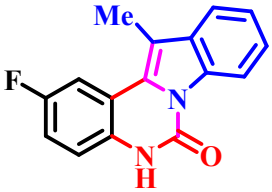
9-Chloro-2,12-dimethylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6bc): The compound **(6bc)** was prepared using 5-methylisatin (0.50 mmol) and 5-chloro-2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound aswhite solid (104.0 mg, 0.35 mmol, 70% yield), **M.P.** 286-288 ^{o}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.21- 11.19 (m, 1H), 8.48-8.46 (m, 1H), 7.77-7.67 (m, 2H), 7.34-7.30 (m, 1H), 7.17-7.06 (m, 2H), 2.57 (s, 3H), 2.36 (s, 3H); **^{13}C NMR** (100 MHz, , d_6-DMSO): δ 147.3, 132.7, 132.6, 132.4, 129.9, 129.7, 127.9, 124.4, 123.6, 120.0, 115.7, 115.4, 115.1, 108.8, 21.2, 10.8; **FT-IR** (KBr, cm^{-1}): υ_{max}1066, 1135, 1239, 1335, 1372, 1412, 1495, 1596, 1643, 2926, 3082, 3213, 3422; ESI-MS (*m*/*z*) for $C_{17}H_{14}ClN_2O$ $[M+H]^+$: Calculated 297.0795, found 297.0799.



2-Methoxy-10,12-dimethylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6cb): The compound **(6cb)** was prepared using 5-methoxyisatin(0.50 mmol) and2-ethynyl-4-methylaniline (0.50 mmol) as starting materials. Purification by column chromatography (10% ethyl acetate in petroleum ether) afforded the title compound as off-white solid (108.3 mg, 0.37 mmol, 74% yield), **M.P.** 264-266 ^{o}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.05 (s, 1H), 8.41 (d, *J*=8.4Hz, 1H), 7.51-7.49 (m, 2H), 7.19-7.15 (m, 2H), 7.02-7.00 (m, 1H), 3.84(s, 3H), 2.61(s, 3H), 2.46(s, 3H); **^{13}C-NMR** (100 MHz, , d_6-DMSO): δ 155.2, 147.4, 132.4, 131.3, 131.0, 128.9, 128.7, 125.3, 118.3, 116.7, 116.2, 115.7, 115.6, 109.0, 108.2, 55.8, 21.7, 10.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1068, 1137, 1245, 1337, 1370, 1417, 1492, 1599, 1647, 2923, 3078, 3212, 3418; ESI-MS (*m*/*z*) for $C_{18}H_{17}N_2O_2$ [M+H]$^{+}$: Calculated 293.1290, found 293.1293.

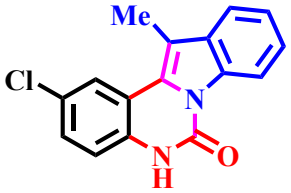


12-Methyl-2-nitroindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ga): The compound **(6ga)** was prepared using 5-nitroisatin (0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as yellow solid (117.2 mg, 0.40 mmol, 80% yield), **M.P.** 245-247 ^{o}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.86 (s, 1H), 8.78 (d, *J*= 2.4Hz, 1H), 8.54-8.51 (m,1H), 8.28-8.25 (m, 1H), 7.86-7.84 (m,1H), 7.45-7.37 (m, 3H), 2.71 (s, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.0, 142.6, 140.4, 132.7, 130.9, 127.0, 124.9, 124.3, 124.0, 119.9, 119.2, 116.3, 115.9, 115.6, 111.7, 10.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1061, 1129, 1239, 1333, 1366, 1412, 1487, 1596, 1640, 2929, 3087, 3215, 3427; ESI-MS (*m*/*z*) for $C_{16}H_{12}N_3O_3$ [M+H]$^{+}$: Calculated 294.0879, found 294.0877.

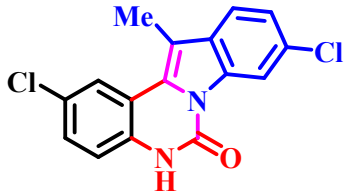


2-Fluoro-12-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one(6da): The compound **(6da)** was prepared using 5-fluoroisatin(0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as off white solid(106.6 mg, 0.40 mmol, 80% yield), **M.P.** 280-284 ^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.25 (s, 1H), 8.56-8.53 (m,1H), 7.77 (d, *J*= 8.4 Hz, 2H), 7.39-7.36 (m, 2H),

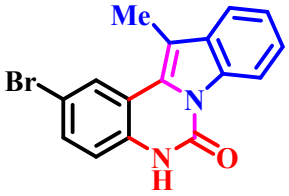
7.25 (d, *J*= 6.6 Hz, 2H), 2.63 (s, 3H); **^{13}C-NMR** (100 MHz,d_6-DMSO): δ 158.1 (C-F, $^{1}J_{C\text{-}F}$= 237 Hz), 147.3, 132.7, 131.4, 130.9, 127.9, 127.9, 124.2, 123.6, 118.9, 117.2 (C-F, $^{3}J_{C\text{-}F}$= 9 Hz), 116.4 (C-F, $^{3}J_{C\text{-}F}$= 9 Hz), 116.0 (C-F, $^{2}J_{C\text{-}F}$= 22 Hz), 110.5, 110.3 (C-F, $^{2}J_{C\text{-}F}$= 25 Hz), 10.6; **FT-IR** (KBr, cm^{-1}): υ_{max}1063, 1131, 1239, 1334, 1368, 1412, 1489, 1598, 1642, 2926, 3087, 3214, 3425; ESI-MS (*m*/*z*) for $C_{16}H_{12}FN_2O$ $[M+H]^+$: Calculated 267.0934, found 267.0936.



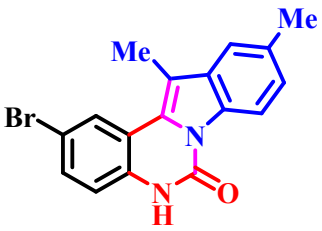
2-Chloro-12-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ea): The compound **(6ea)** was prepared using 5-chloroisatin (0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (113.2 mg, 0.40 mmol, 80% yield), **M.P.** 310-312 °C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.38 (s, 1H), 8.55-8.53 (m, 1H), 7.98 (d,*J* = 2.4 Hz, 1H), 7.81-7.79 (m, 1H), 7.46-7.38 (m, 3H), 7.26(d, *J*= 8.8 Hz, 1H), 2.65 (s, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.3, 133.8, 132.7, 131.0, 128.6, 127.6, 127.1, 124.4, 123.7, 123.6, 119.0, 117.4, 117.0, 115.9, 110.6, 10.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1064, 1132, 1238, 1334, 1370, 1412, 1491, 1596, 1644, 2925, 3082, 3214, 3423; ESI-MS (*m*/*z*) for $C_{16}H_{12}ClN_2O$ $[M+H]^+$: Calculated 283.0638, found 283.0635.



2,9-Dichloro-12-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6ec): The compound **(6ec)** was prepared using 5-chloroisatin (0.50 mmol) and 5-chloro-2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as white solid (122.1 mg, 0.385 mmol, 77% yield), **M.P.** 300-302 °C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.44 (s, 1H), 8.48 (d,*J* = 1.8 Hz, 1H), 7.91 (d, J = 2.4 Hz, 1H), 7.78 (d, *J*= 8.4 Hz, 1H), 7.45-7.36 (m, 2H), 7.22 (d,*J*= 8.7 Hz, 1H), 2.60 (s, 3H); **^{13}C NMR** (75 MHz, d_6-DMSO): δ 146.8, 133.6, 132.6, 129.5, 128.6, 128.5, 128.2, 127.2, 123.7, 123.3, 120.0, 117.3, 116.4, 115.3, 110.1, 10.5; **FT-IR** (KBr, cm^{-1}): υ_{max}1064, 1131, 1236, 1334, 1369, 1412, 1490, 1594, 1639, 2925, 3082, 3215, 3425; ESI-MS (*m*/*z*) for $C_{16}H_{11}Cl_2N_2O$ $[M+H]^+$: Calculated 317.0248, found 317.0244.



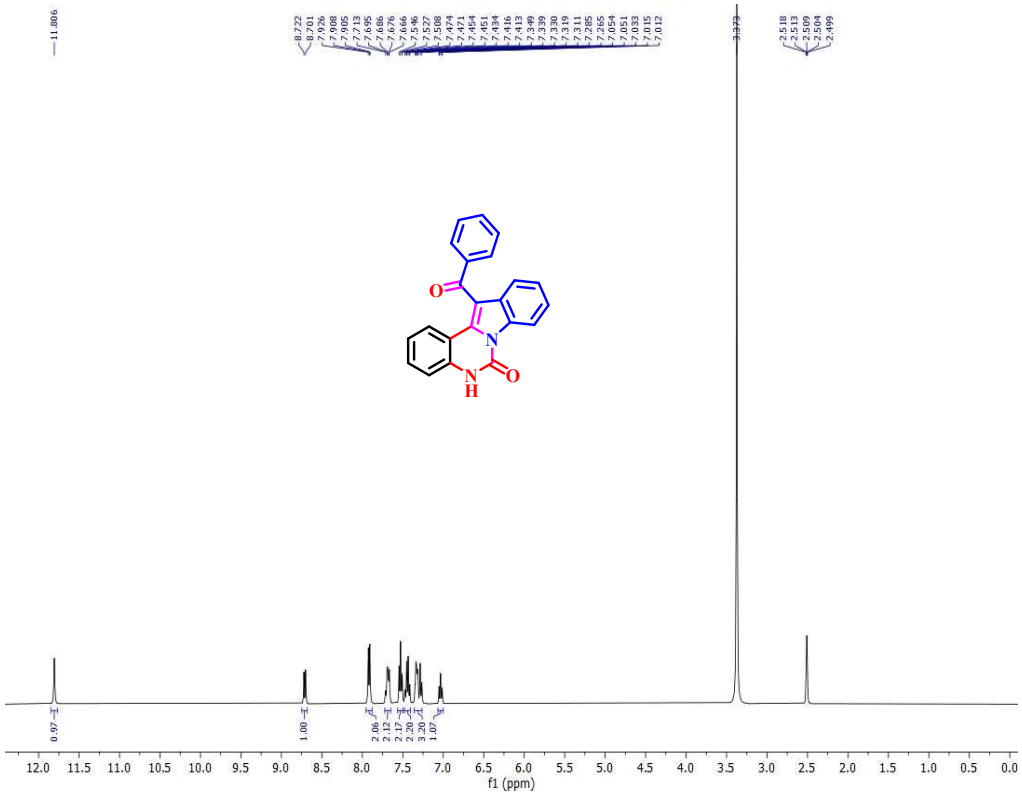
2-Bromo-12-methylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6fa): The compound **(6fa)** was prepared using 5-bromoisatin (0.50 mmol) and 2-ethynylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as light brown solid (130.9 mg, 0.40 mmol, 80% yield), **M.P.** 305-307 ^{o}C; **^{1}H NMR** (300 MHz, d_6-DMSO): δ 11.32 (s, 1H), 8.51-8.48 (m, 1H), 8.02 (d, J = 2.3 Hz, 1H), 7.76-7.73 (m, 1H), 7.50 (dd, *J*= 8.4 Hz, 2.1 Hz,1H), 7.38-7.32 (m, 2H), 7.15 (d, *J*= 8.7Hz, 1H), 2.58 (s, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.3, 134.2, 132.7, 131.4, 131.0, 127.4, 126.4, 124.4, 123.7, 118.9, 117.7, 117.4, 115.9, 114.9, 110.6, 10.7; **FT-IR** (KBr, cm^{-1}): υ_{max}1065, 1132, 1239, 1334, 1371, 1412, 1492, 1596, 1641, 2925, 3080, 3214, 3422;ESI-MS (*m*/*z*) for $C_{16}H_{12}BrN_2O$ $[M+H]^+$: Calculated 327.0133, found 327.0138.

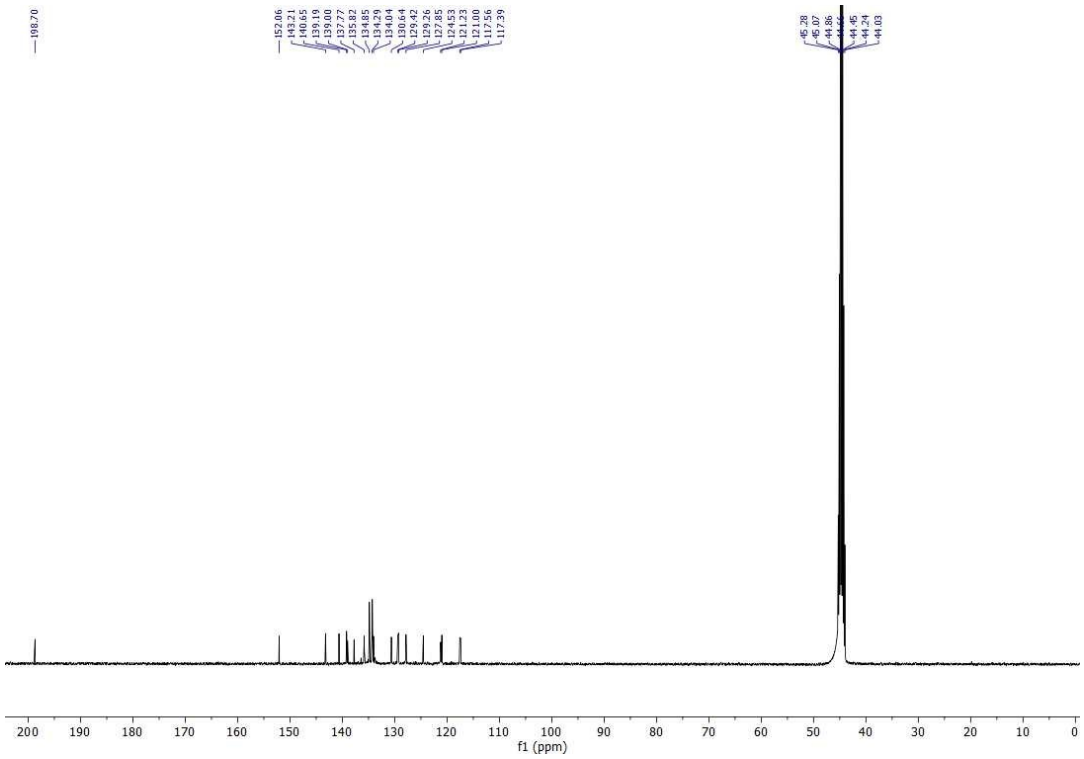


2-Bromo-10,12-dimethylindolo[1,2-*c*]quinazolin-6(5*H*)-one (6fb): The compound **(6fb)** was prepared using 5-bromoisatin (0.50 mmol) and 2-ethynyl-4-methylaniline (0.50 mmol) as starting materials. Purification by column chromatography (5% ethyl acetate in petroleum ether) afforded the title compound as light brown solid (136.5 mg, 0.40 mmol, 80% yield), **M.P.** 296-298 ^{o}C; **^{1}H NMR** (400 MHz, d_6-DMSO): δ 11.24 (s, 1H), 8.32 (d, *J*= 8.4 Hz, 1H), 7.96 (d, *J*= 2.0 Hz, 1H), 7.49-7.45 (m, 2H), 7.14-7.11 (m, 2H), 2.52 (s, 3H), 2.43 (s, 3H); **^{13}C NMR** (100 MHz,d_6-DMSO): δ 147.2, 134.1, 132.8, 131.3, 131.2, 131.0, 127.4, 126.2, 125.9, 118.5, 117.7, 117.5, 115.5, 114.9, 110.3, 21.7, 10.7; **FT-IR** (KBr, cm^{-1}):υ_{max}1066, 1134, 1239, 1336, 1373, 1412, 1492, 1597, 1642, 2925, 3079, 3213, 3421;ESI-MS (*m*/*z*) for $C_{17}H_{14}BrN_2O$ $[M+H]^+$: Calculated 341.0290, found 341.0292.

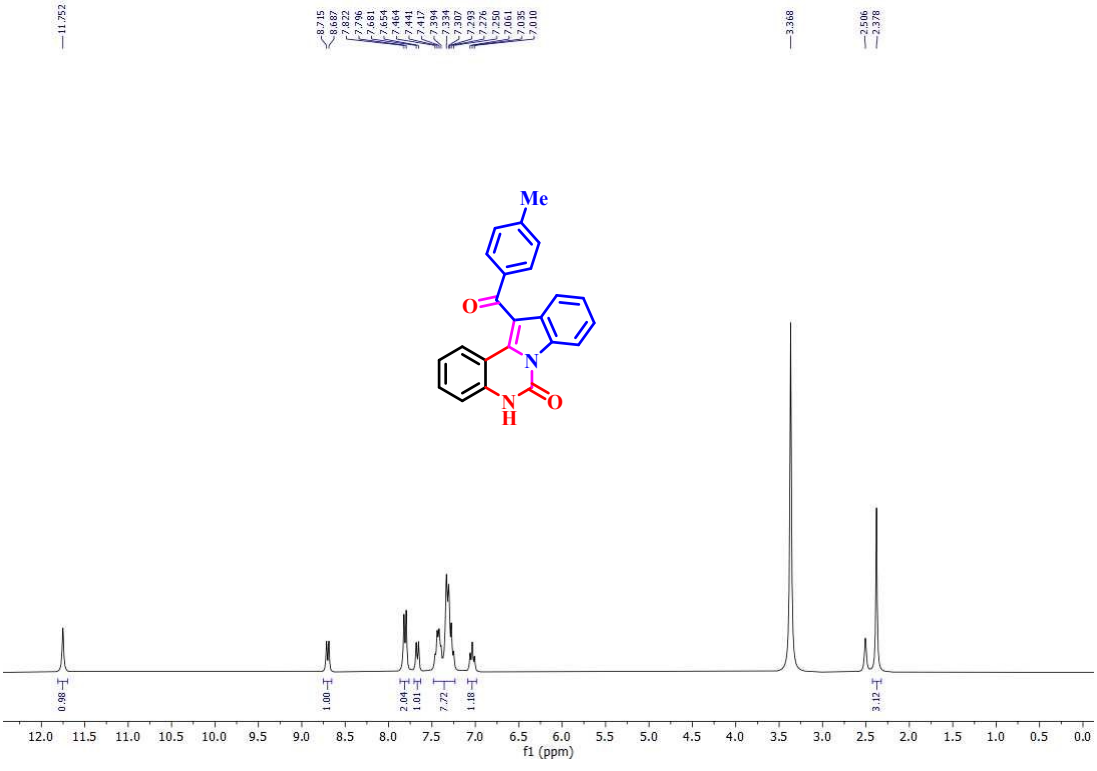
10. NMR Spectra of 12-benzoylindolo[1,2-c]quinazolin-6(5*H*)-one

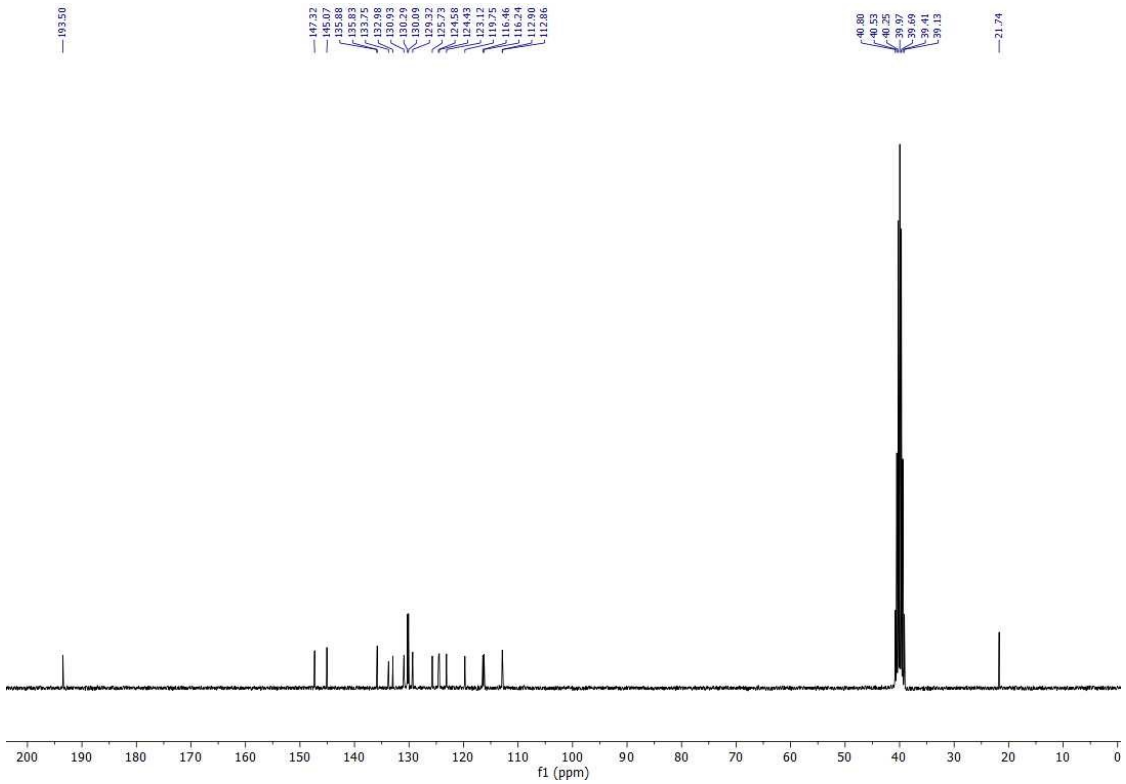
^{1}H and^{13}C-NMR of compound (**4aa**)



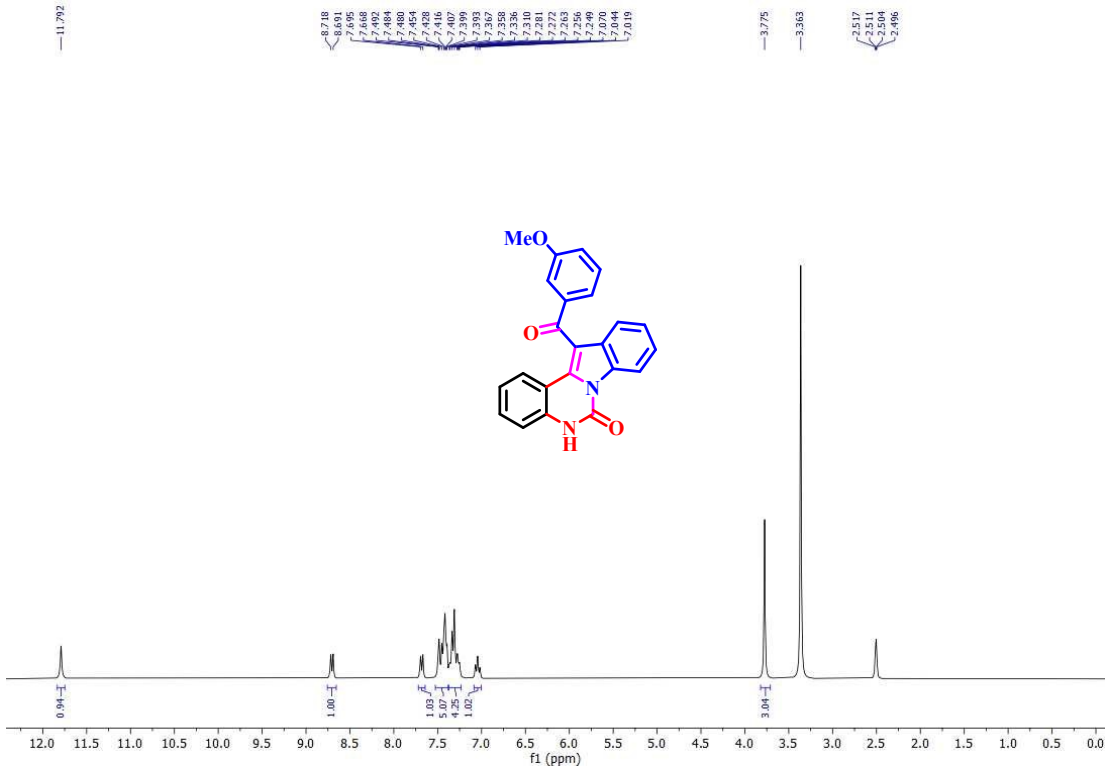


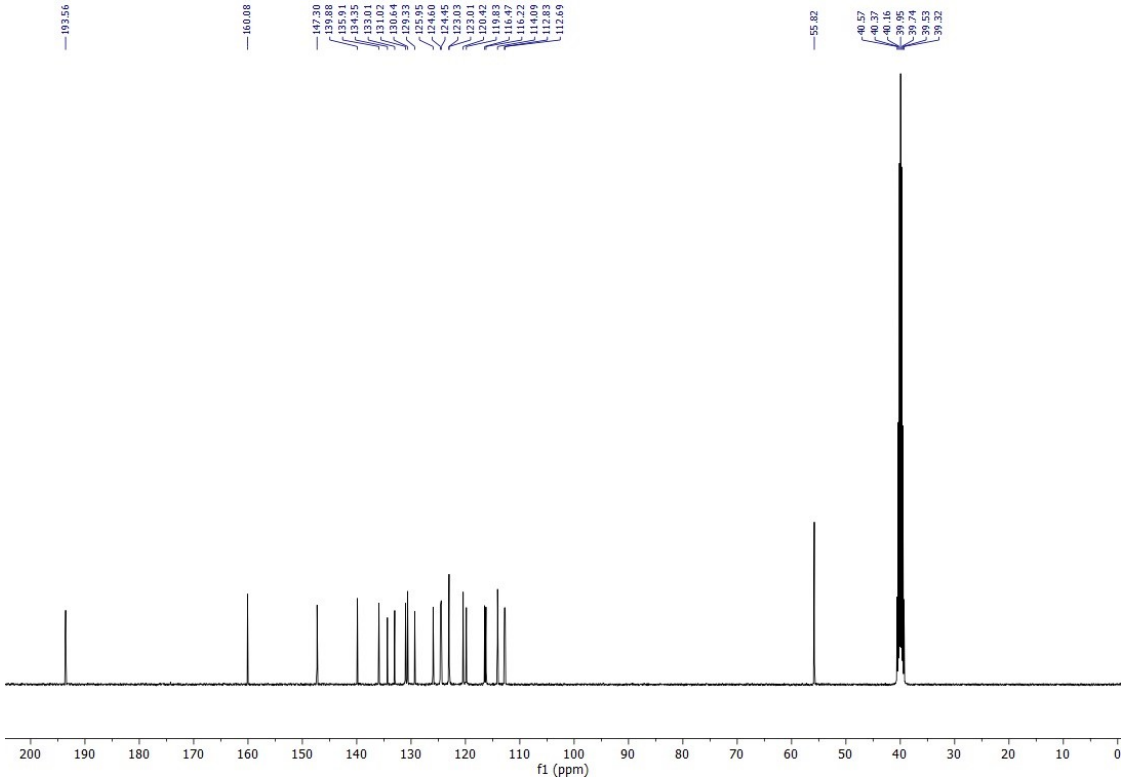
^{1}H and^{13}C-NMR of compound (**4ab**)



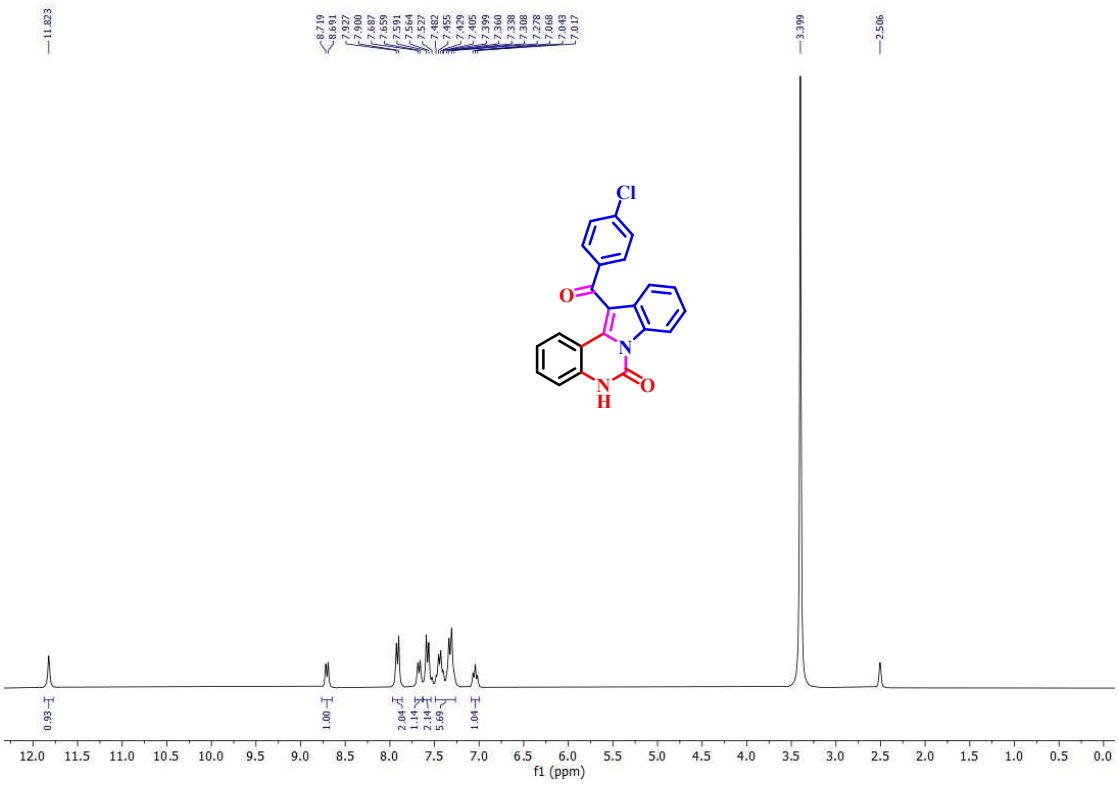


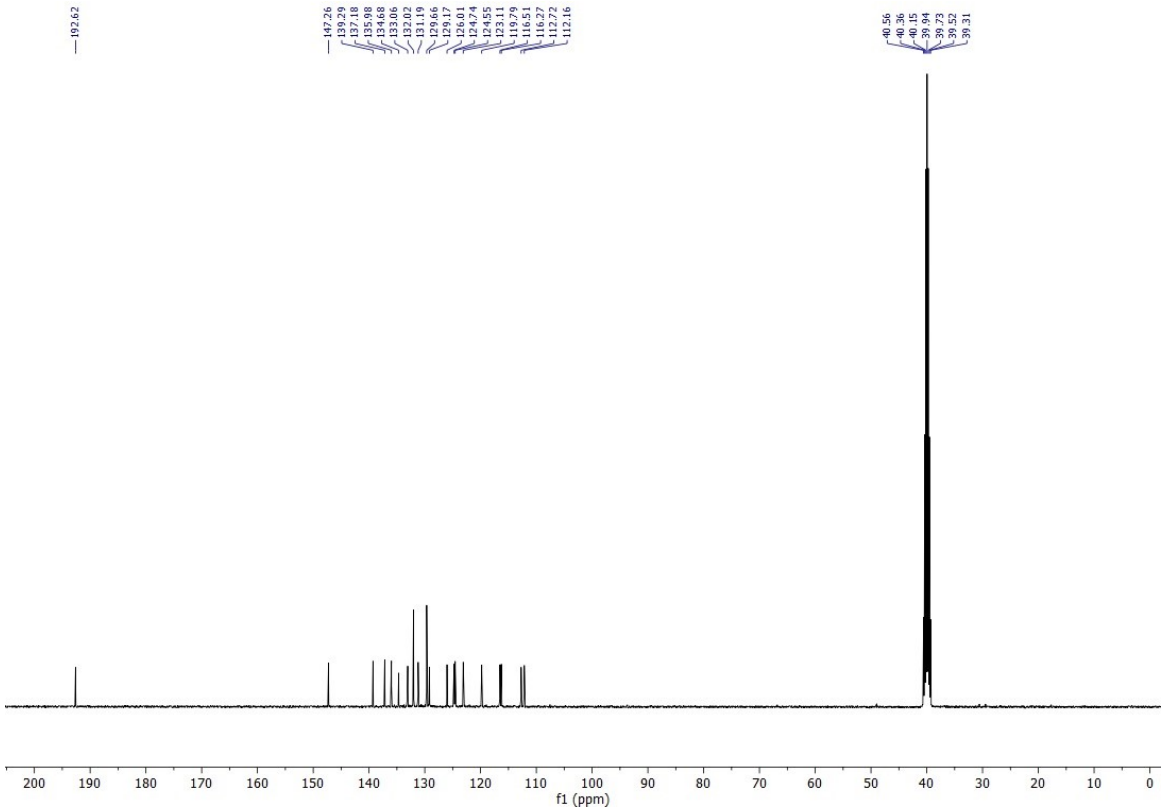
^{1}H and^{13}C-NMR of compound (**4ac**)



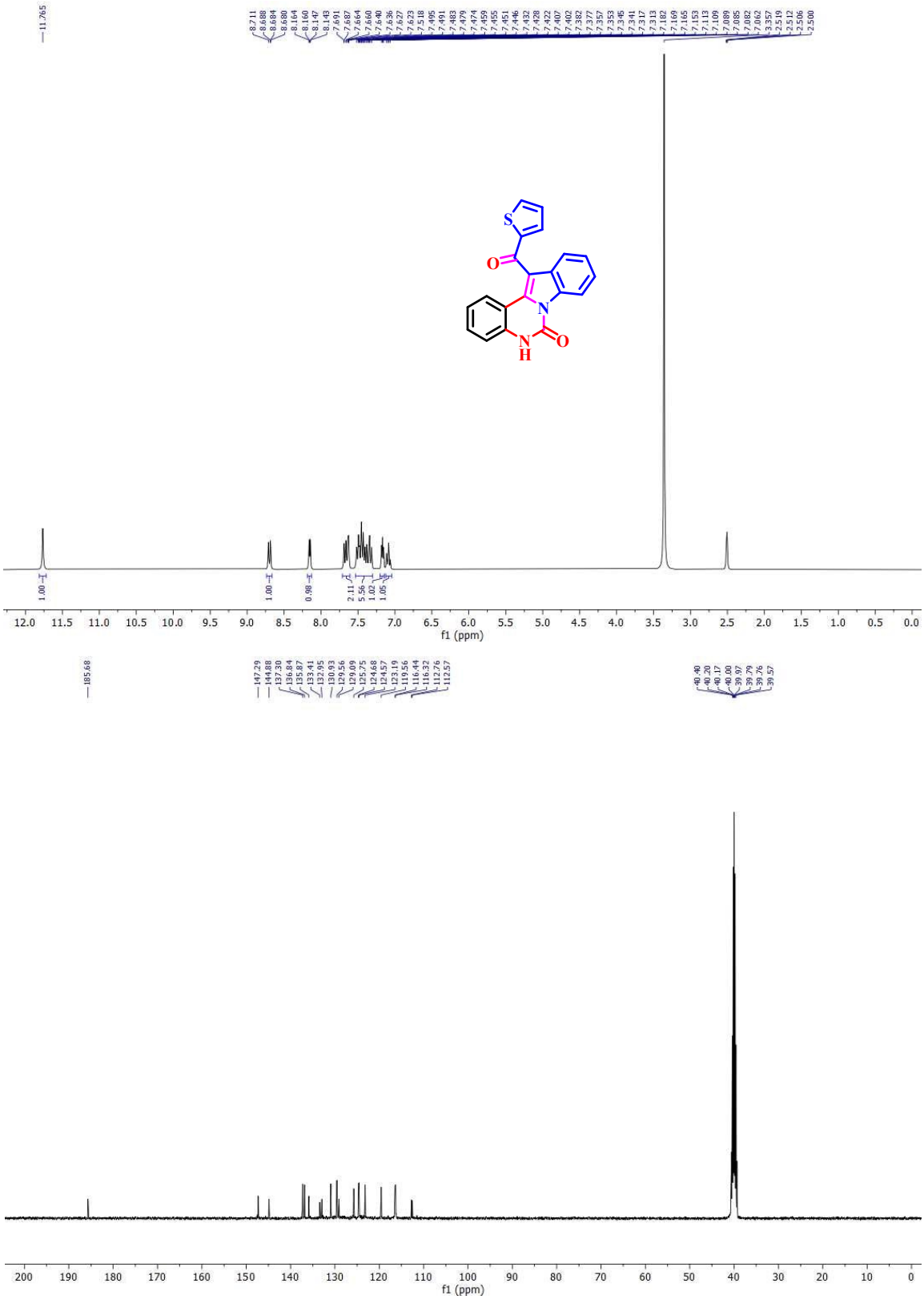


^{1}H and^{13}C-NMR of compound (**4ad**)

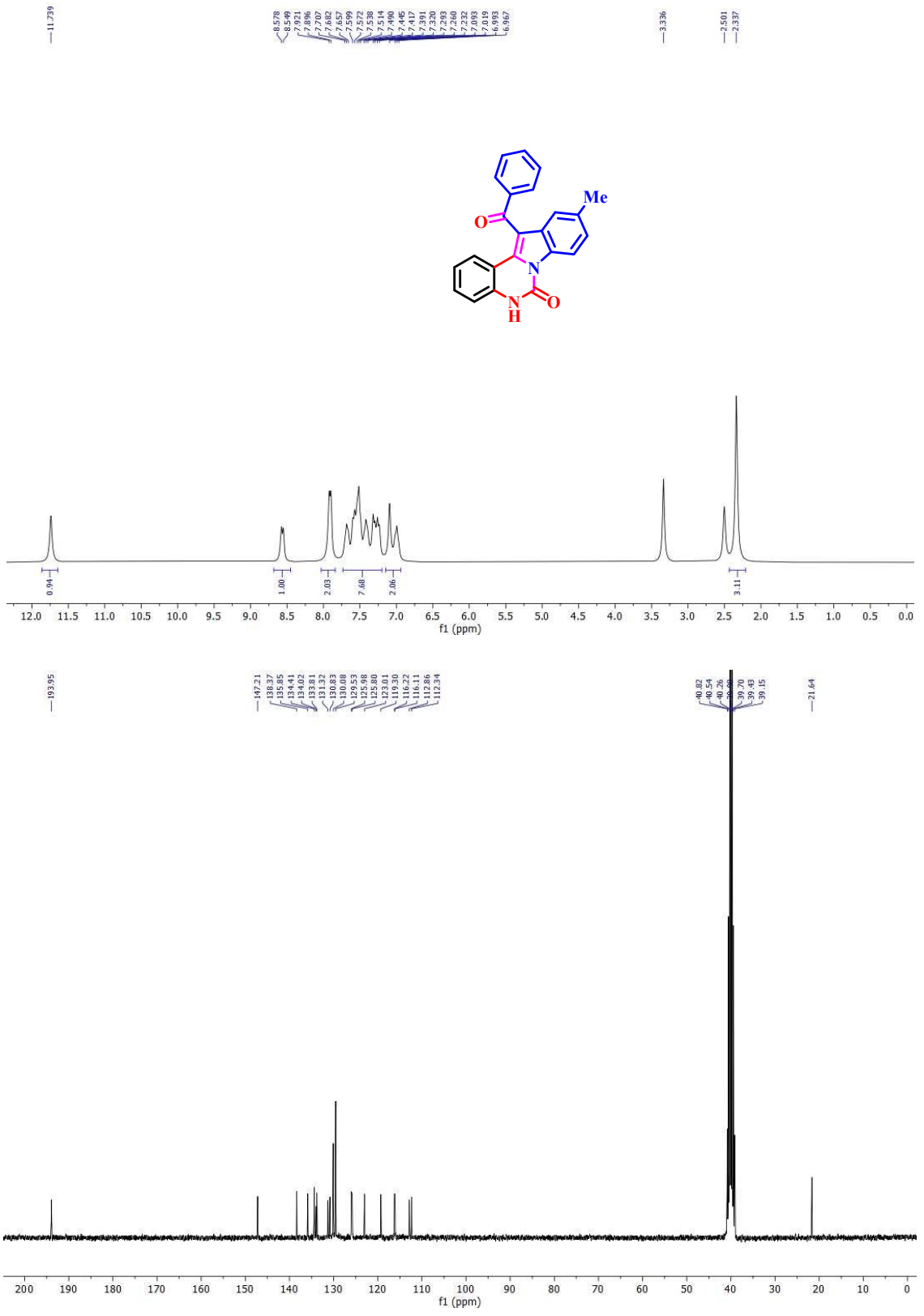




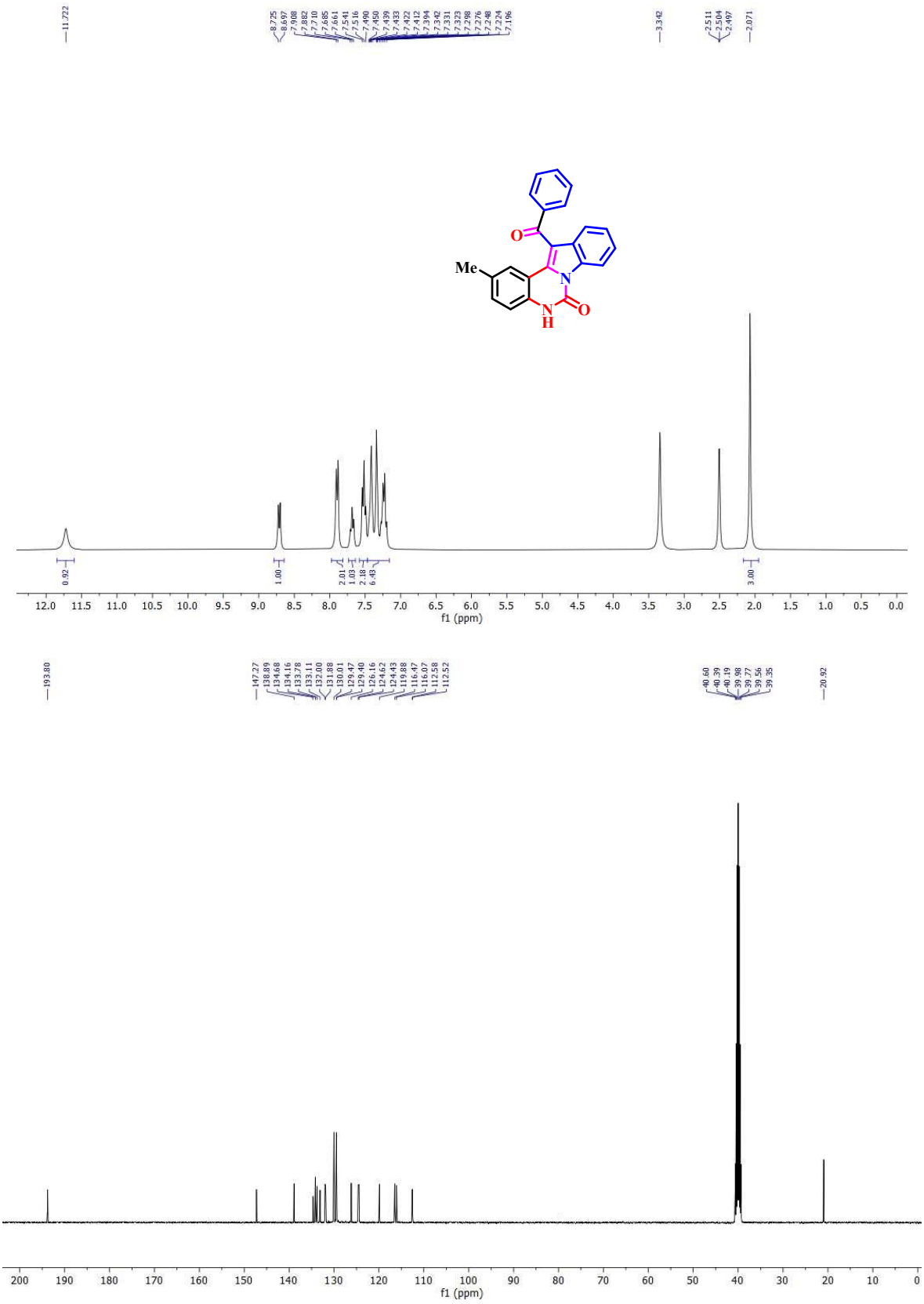
^{1}H and^{13}C-NMR of compound (**4ae**)



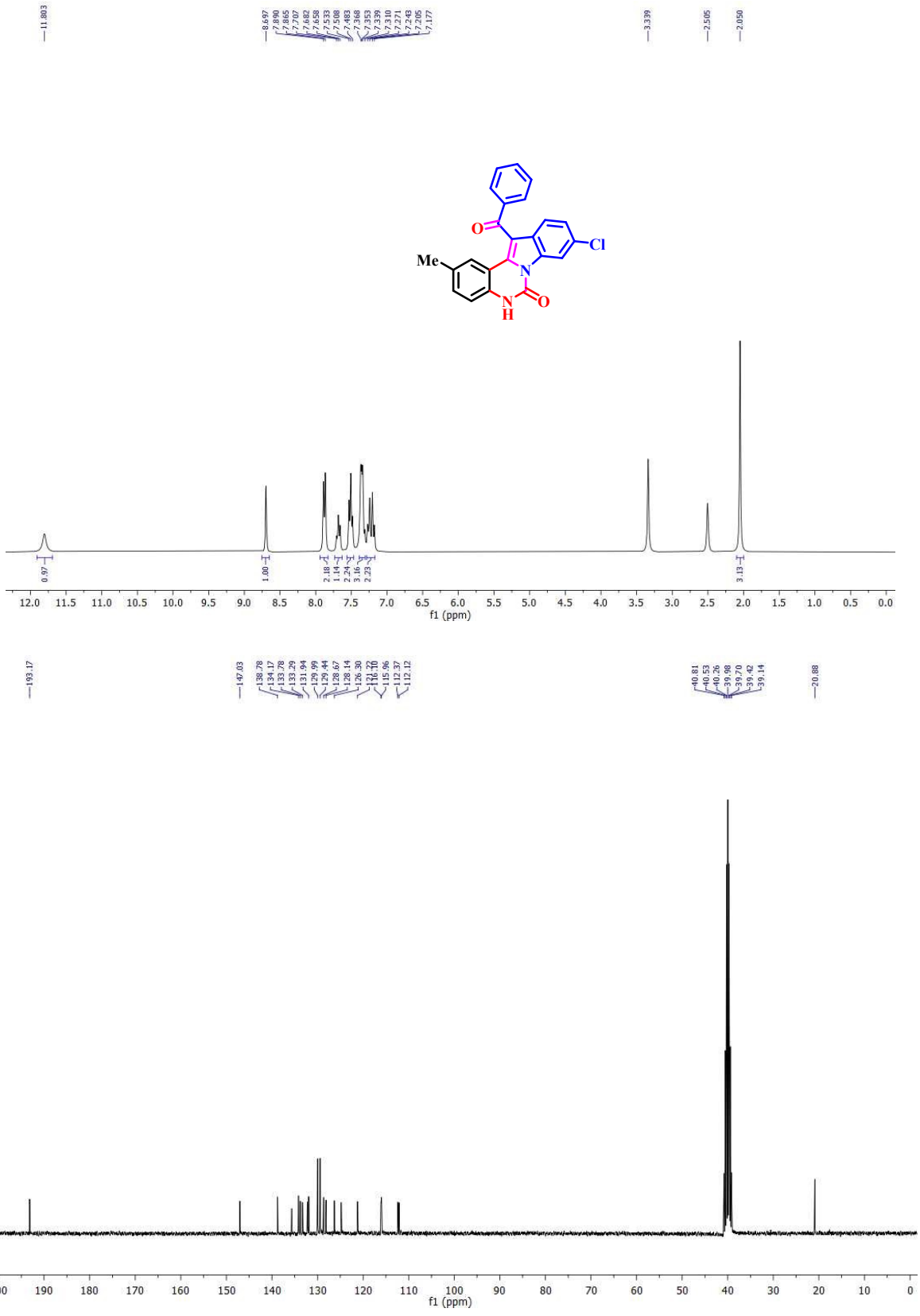
^{1}H and^{13}C-NMR of compound (**4af**)



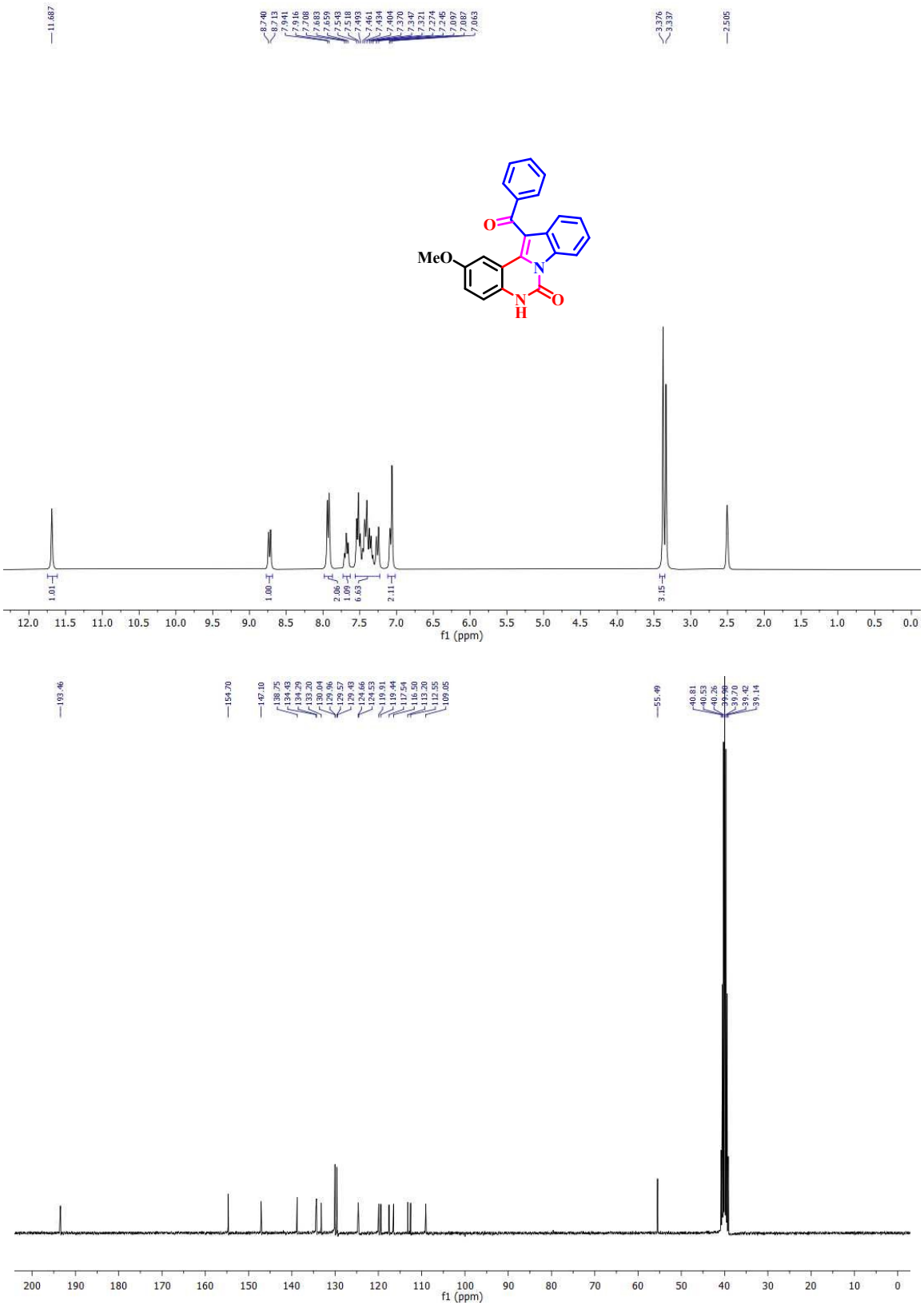
^{1}H and ^{13}C-NMR of compound (**4ba**)



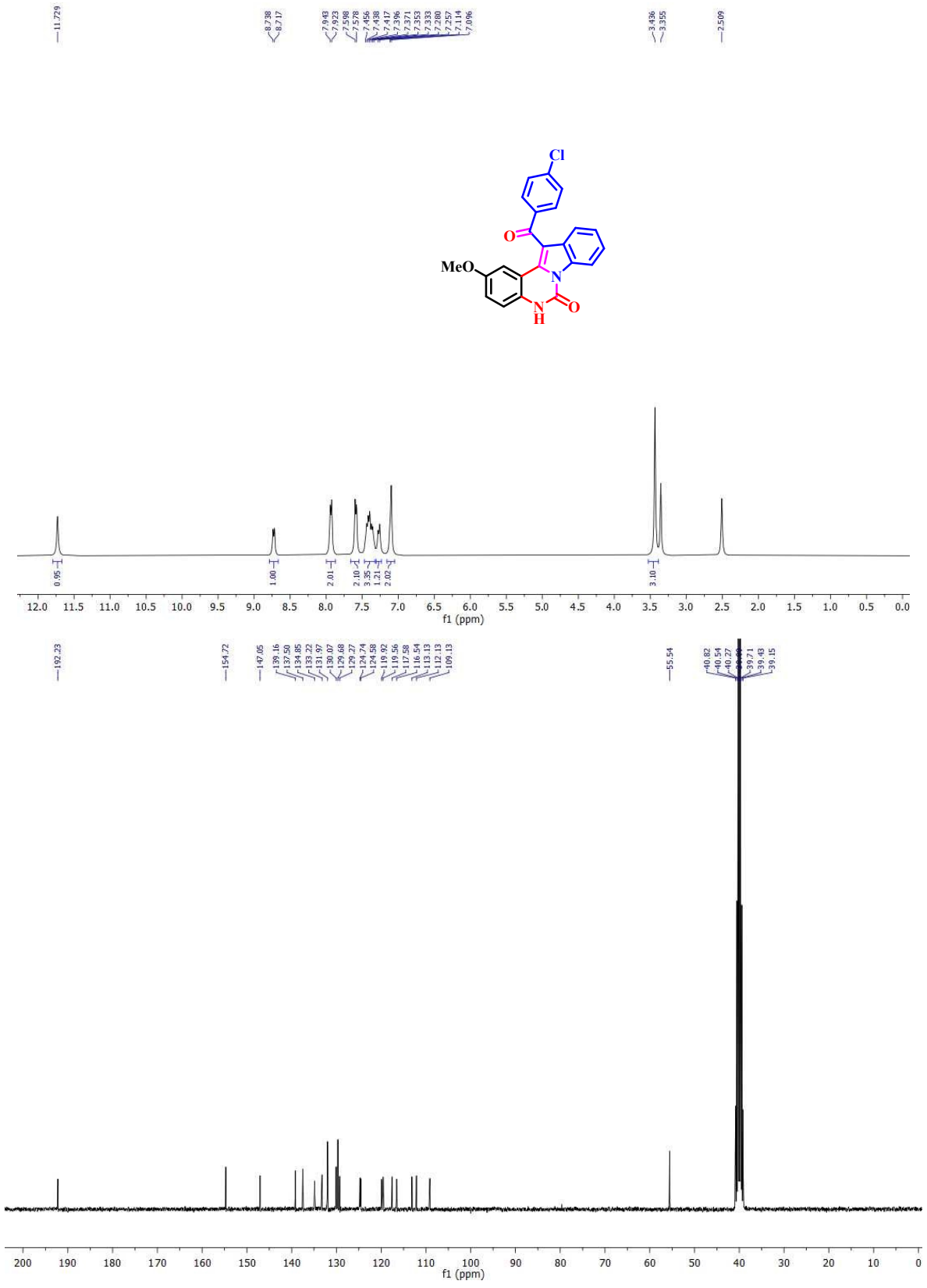
^{1}H and ^{13}C-NMR of compound (**4bg**)



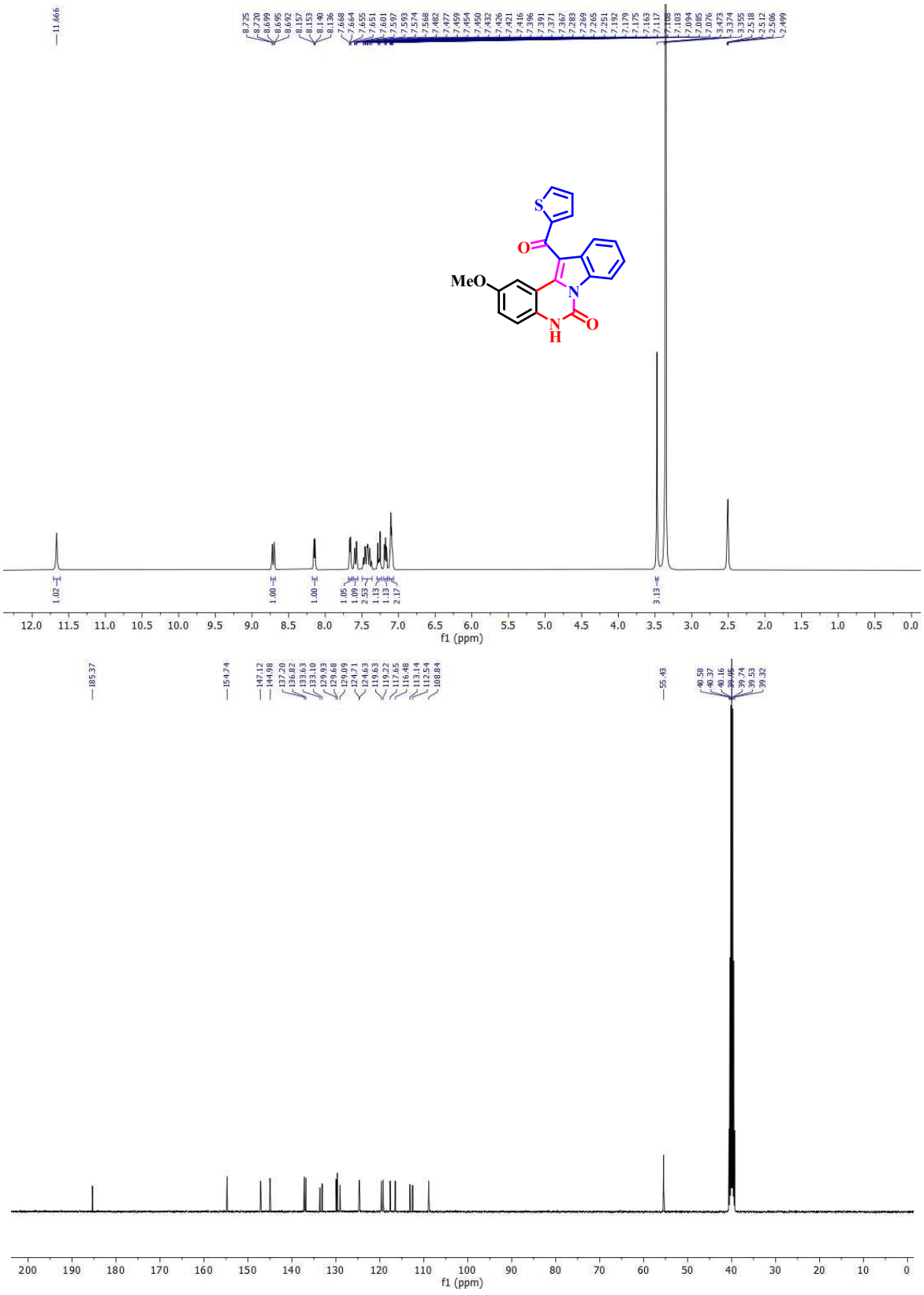
^{1}H and ^{13}C-NMR of compound (**4ca**)



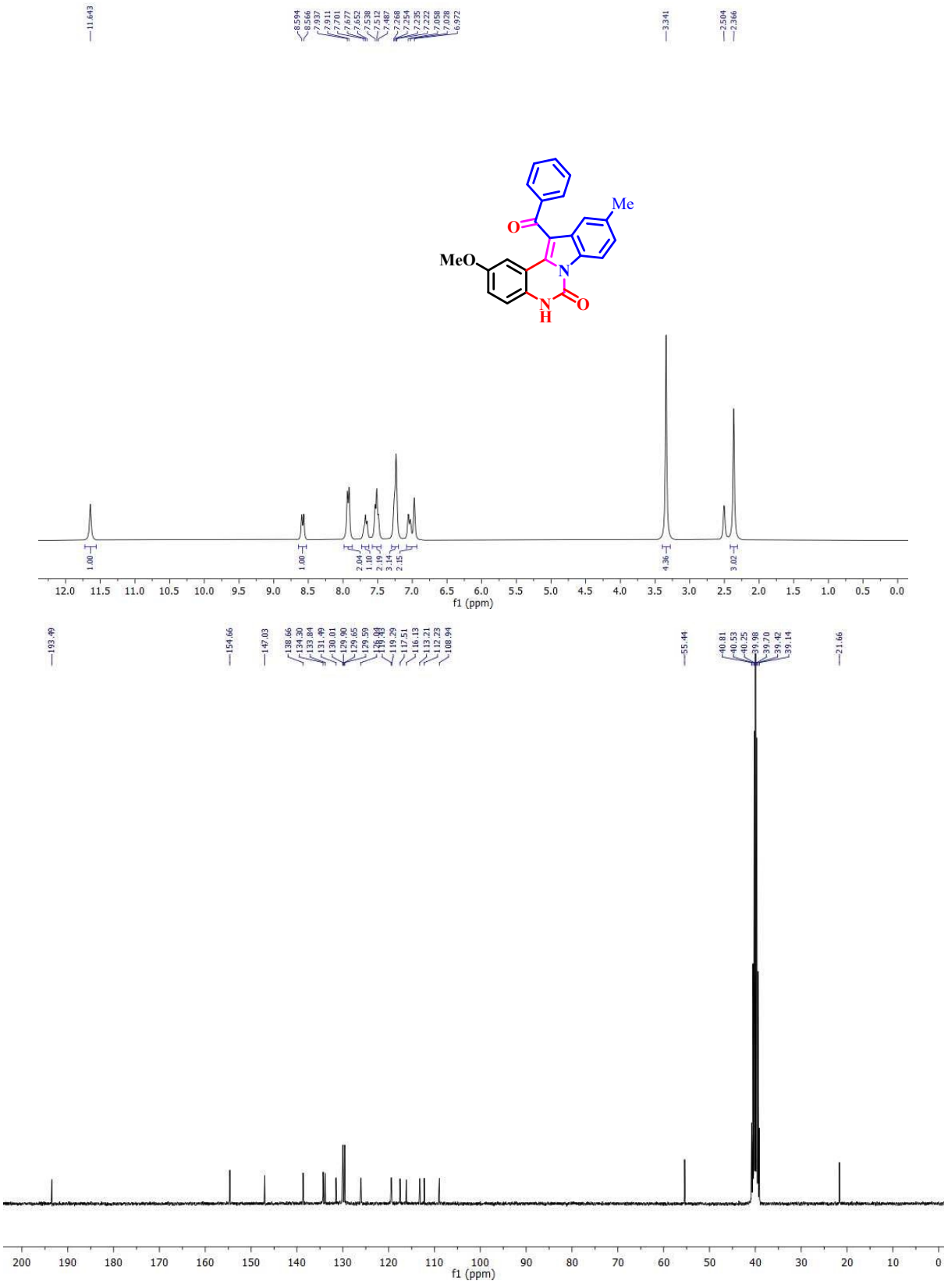
^{1}H and ^{13}C-NMR of compound (**4cd**)



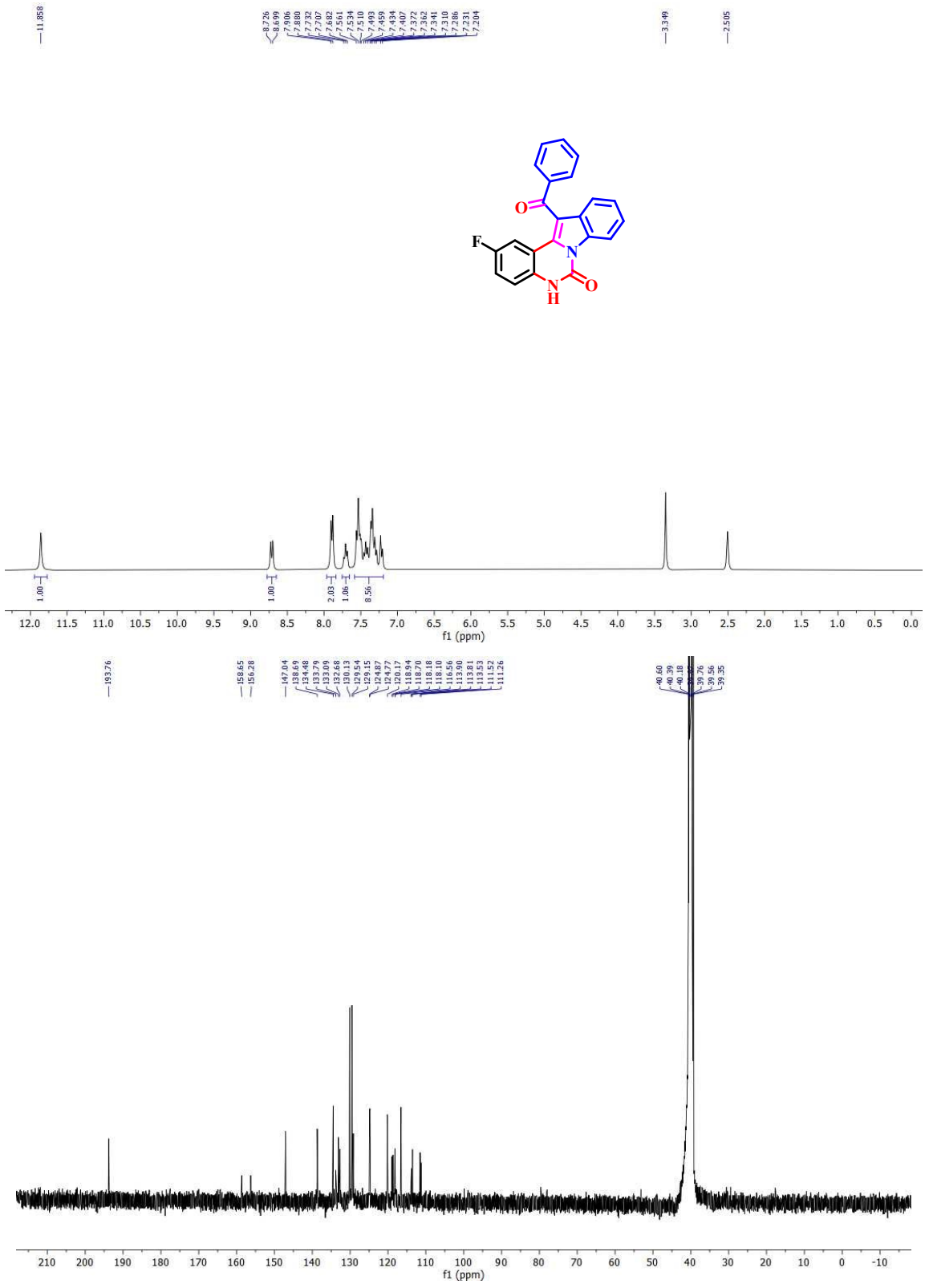
^{1}H and ^{13}C-NMR of compound (**4ce**)



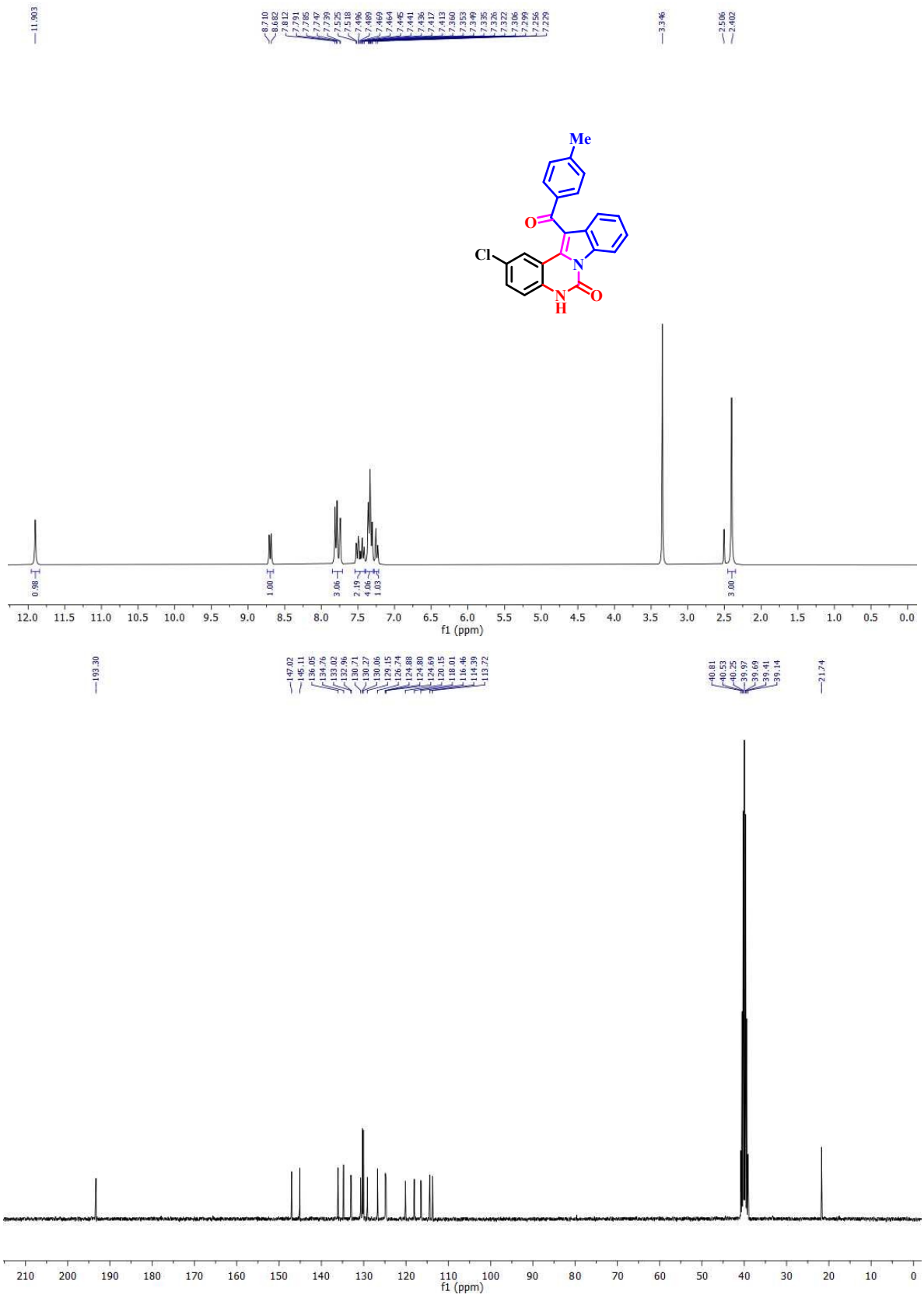
^{1}H and ^{13}C-NMR of compound (**4cf**)



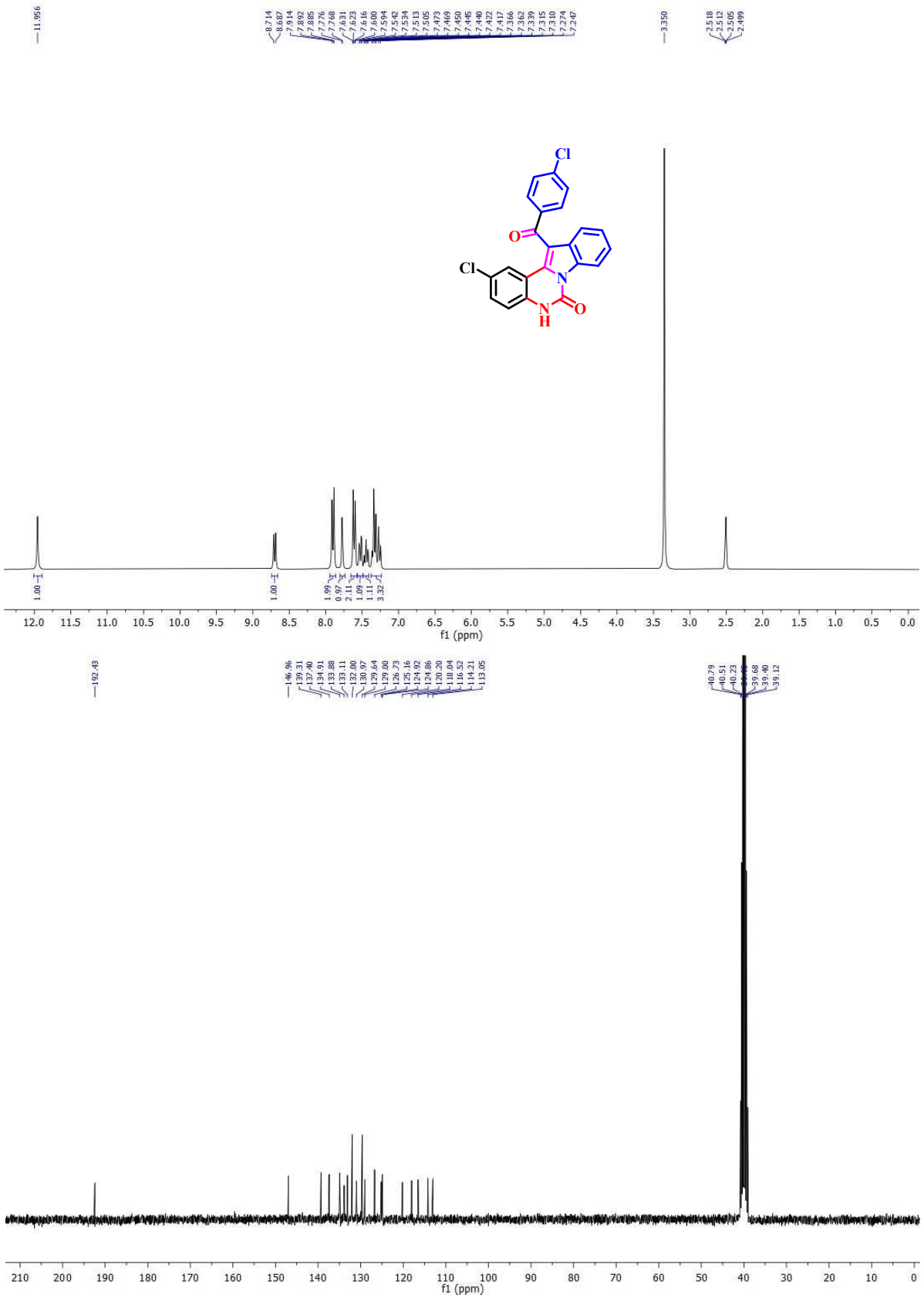
^{1}H and ^{13}C-NMR of compound (**4da**)



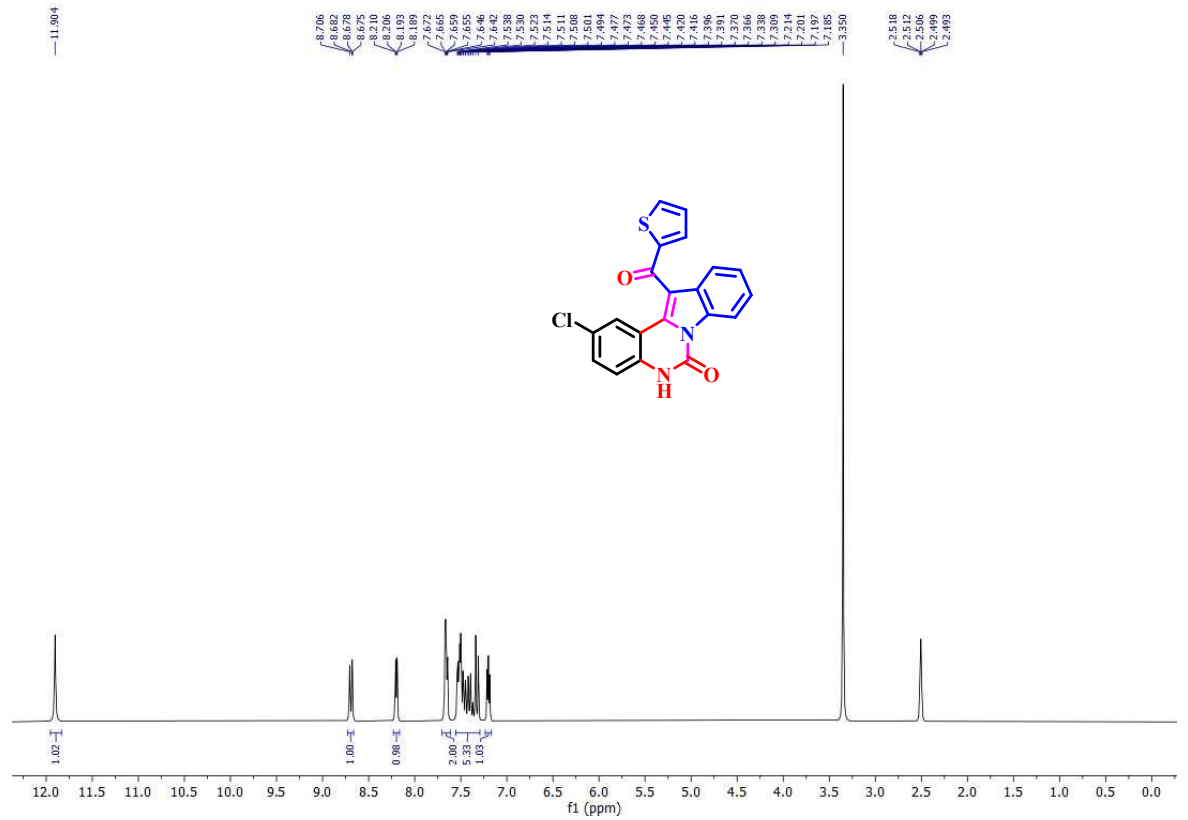
^{1}H and ^{13}C-NMR of compound (**4eb**)



^{1}H and ^{13}C-NMR of compound (**4ed**)



^{1}H and ^{13}C-NMR of compound (**4ee**)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8.682
8.678
8.675
8.210
8.206
8.193
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7.646
7.642
7.538
7.530
7.523
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7.511
7.508
7.501
7.494
7.477
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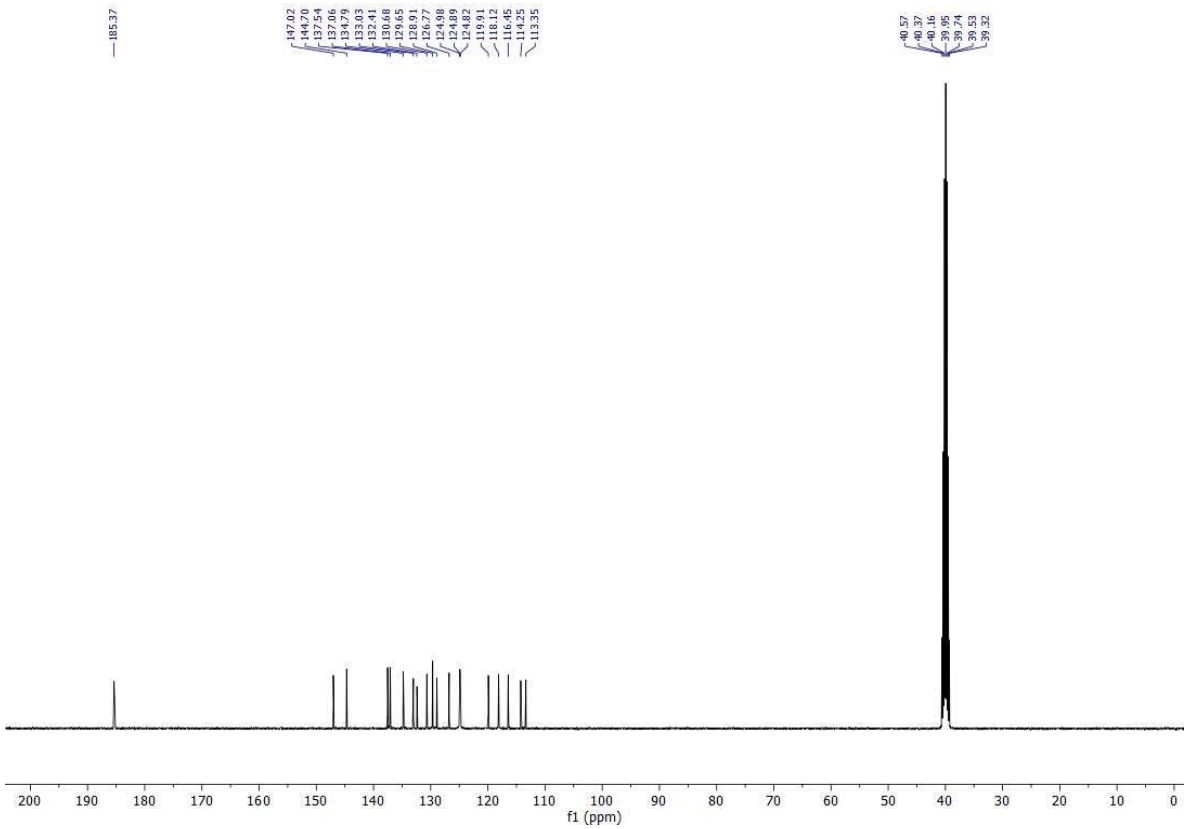
7.445
7.420
7.416
7.396
7.391
7.370
7.366
7.338
7.309
7.214
7.201
7.197
7.185
3.350
2.518
2.512
2.506
2.499
2.493
S
O
Cl
N
N
H
O
1.02
1.00
0.98
2.00
5.33
1.03
12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)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137.06
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129.65
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124.89
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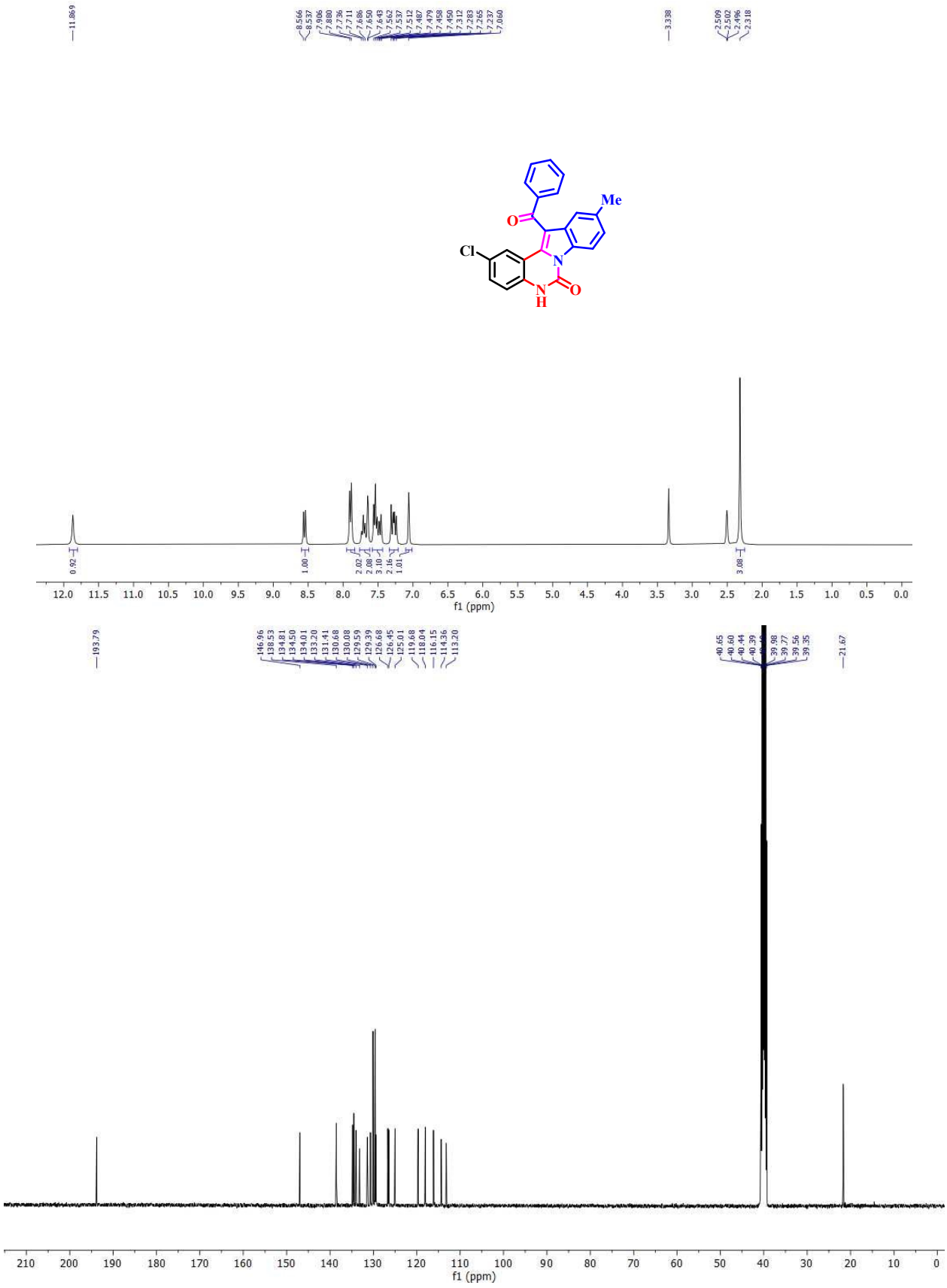
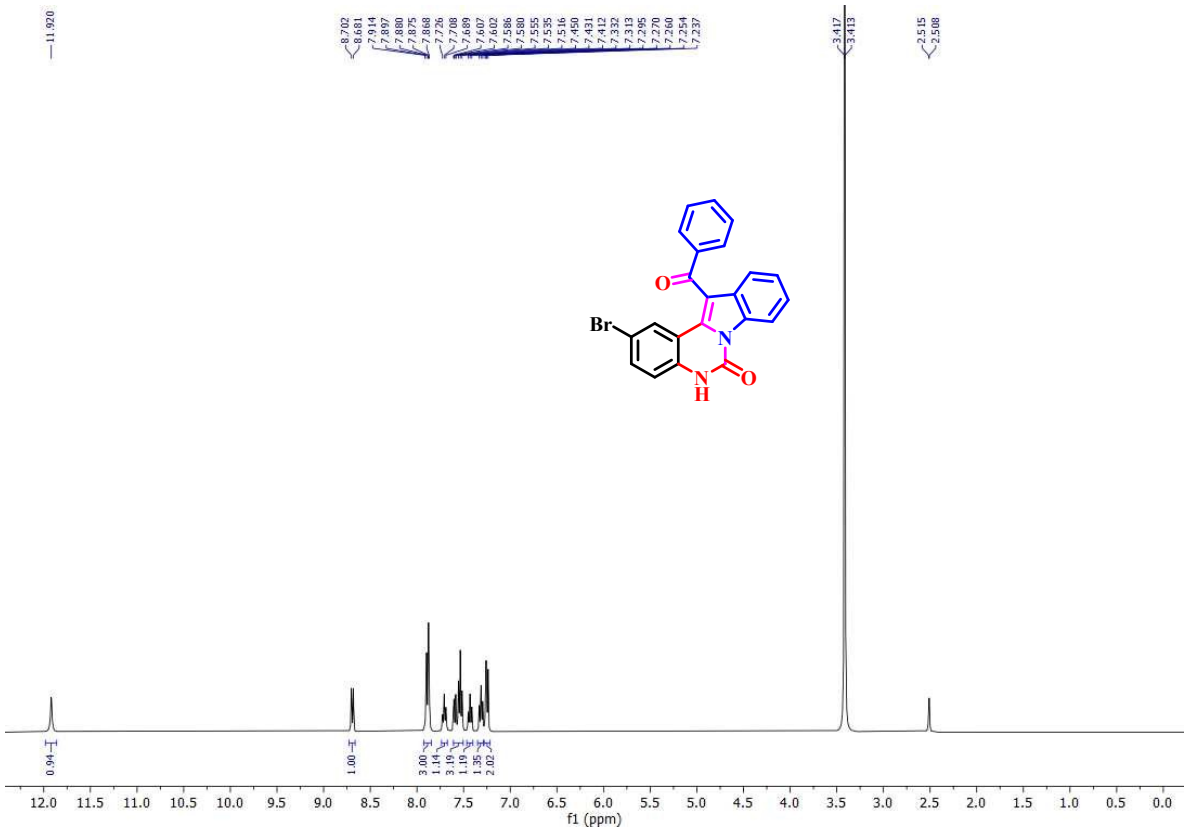
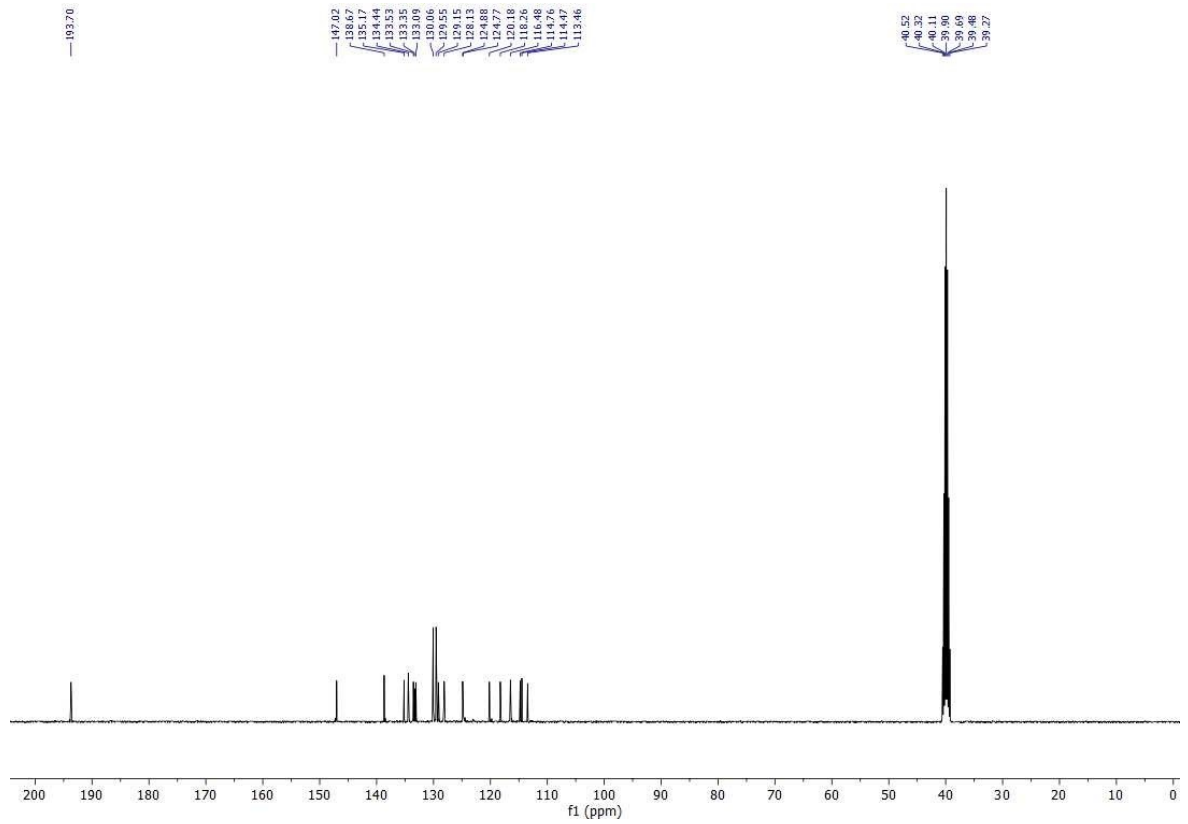
119.91
119.12
116.45
114.25
113.35
40.57
40.37
40.16
39.95
39.74
39.53
39.32
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)

^{1}H and ^{13}C-NMR of compound (**4ef**)

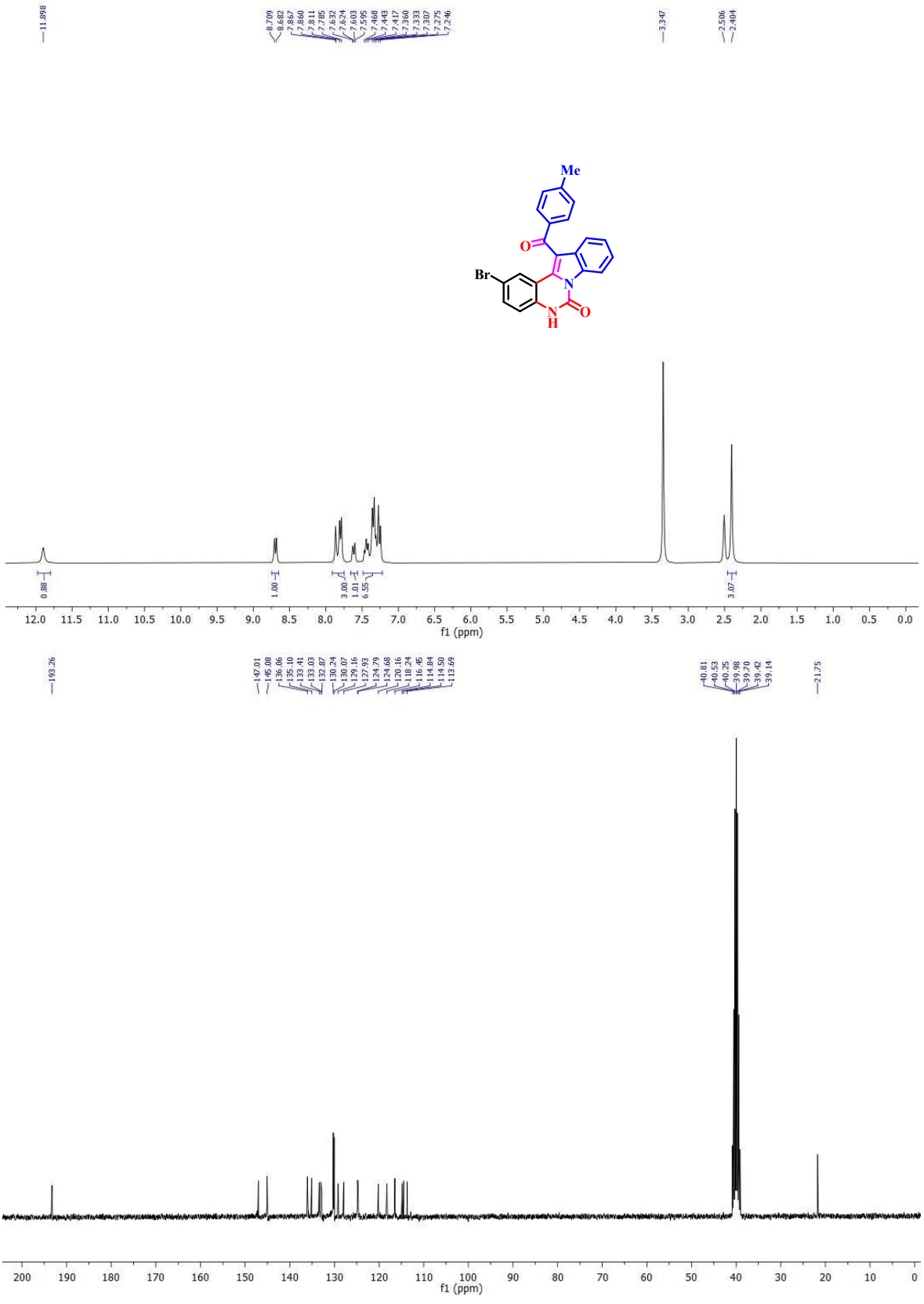


^{1}H and ^{13}C-NMR of compound (**4fa**)

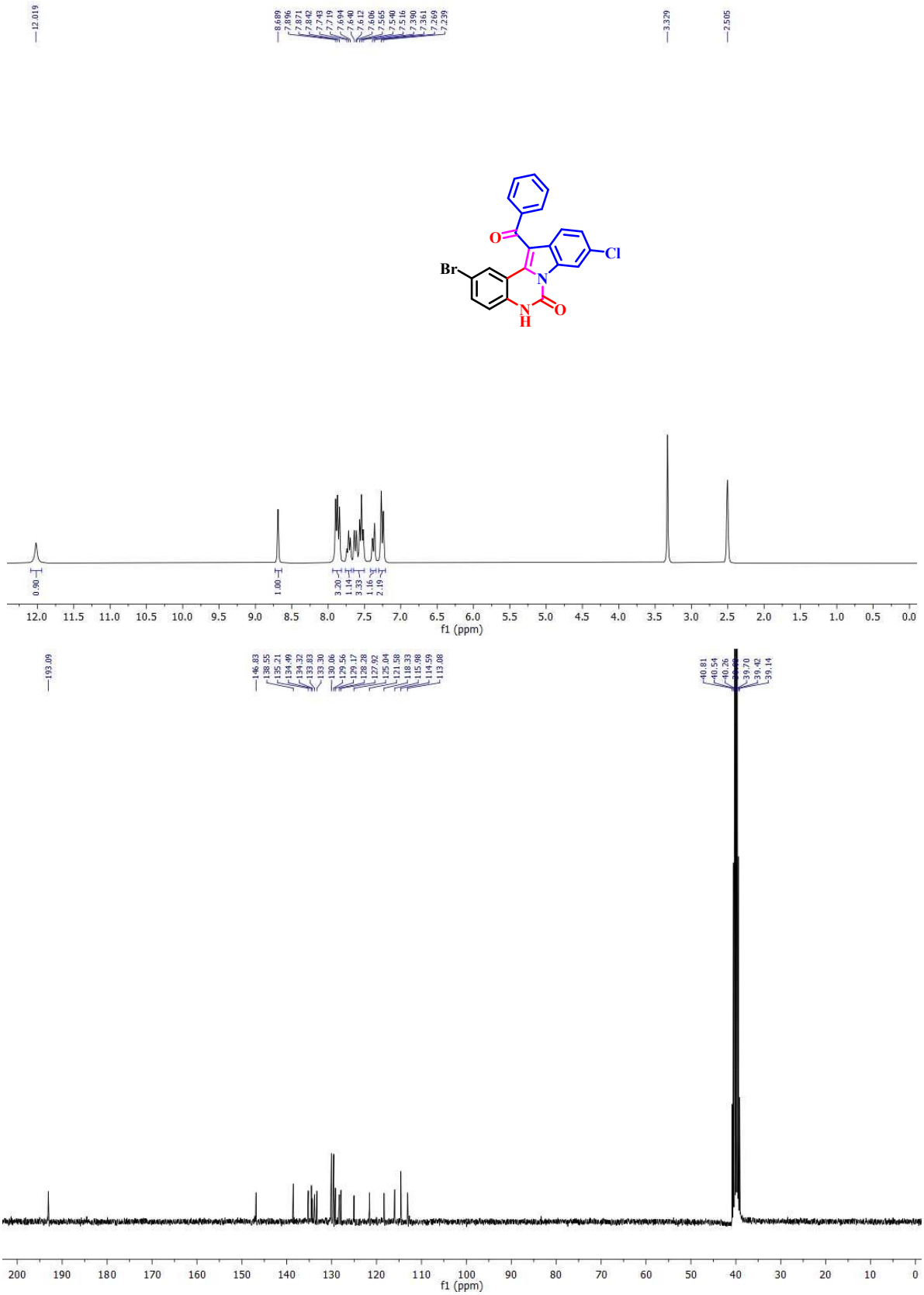




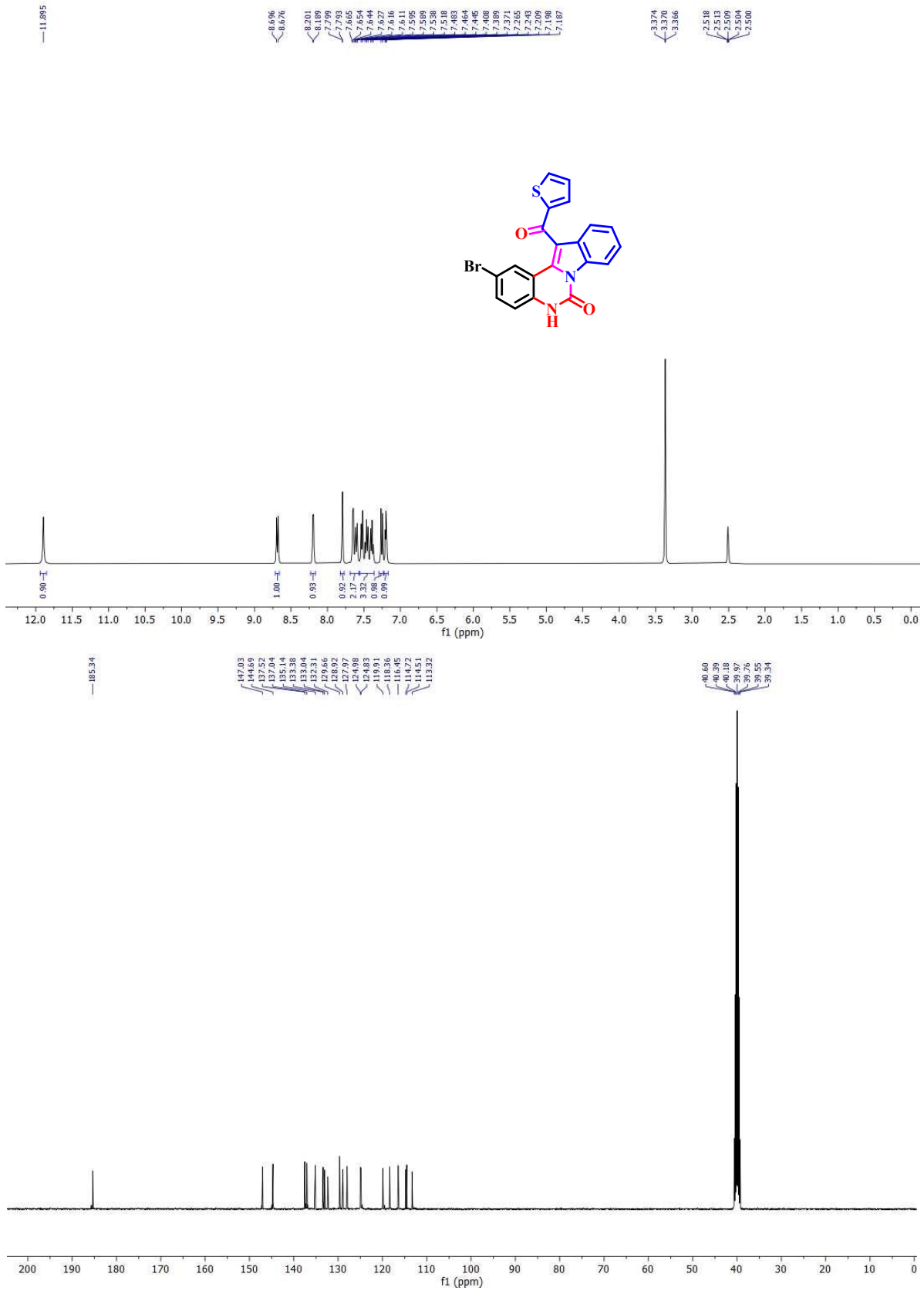
^{1}H and ^{13}C-NMR of compound (**4fb**)



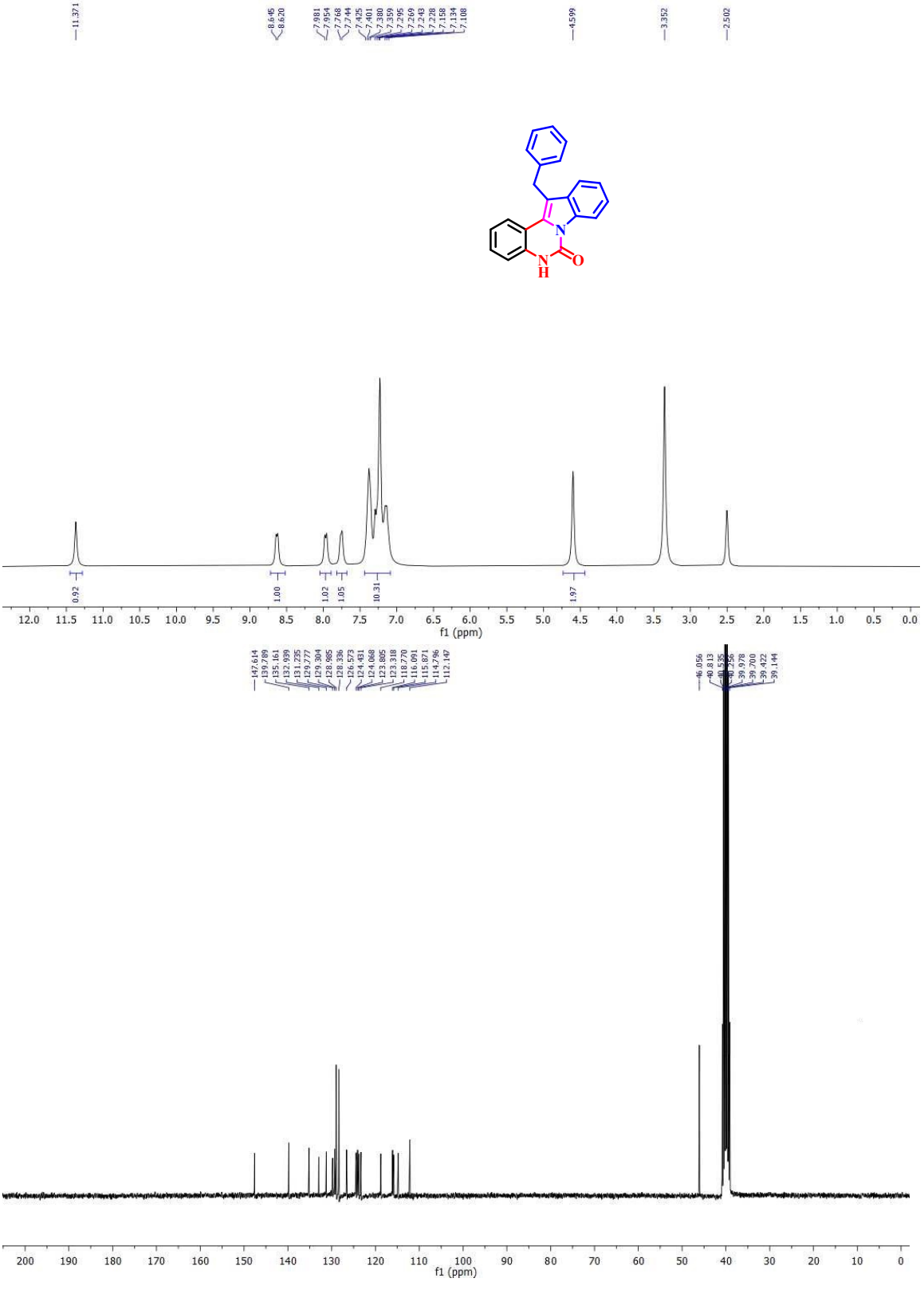
^{1}H and ^{13}C-NMR of compound (**4fg**)



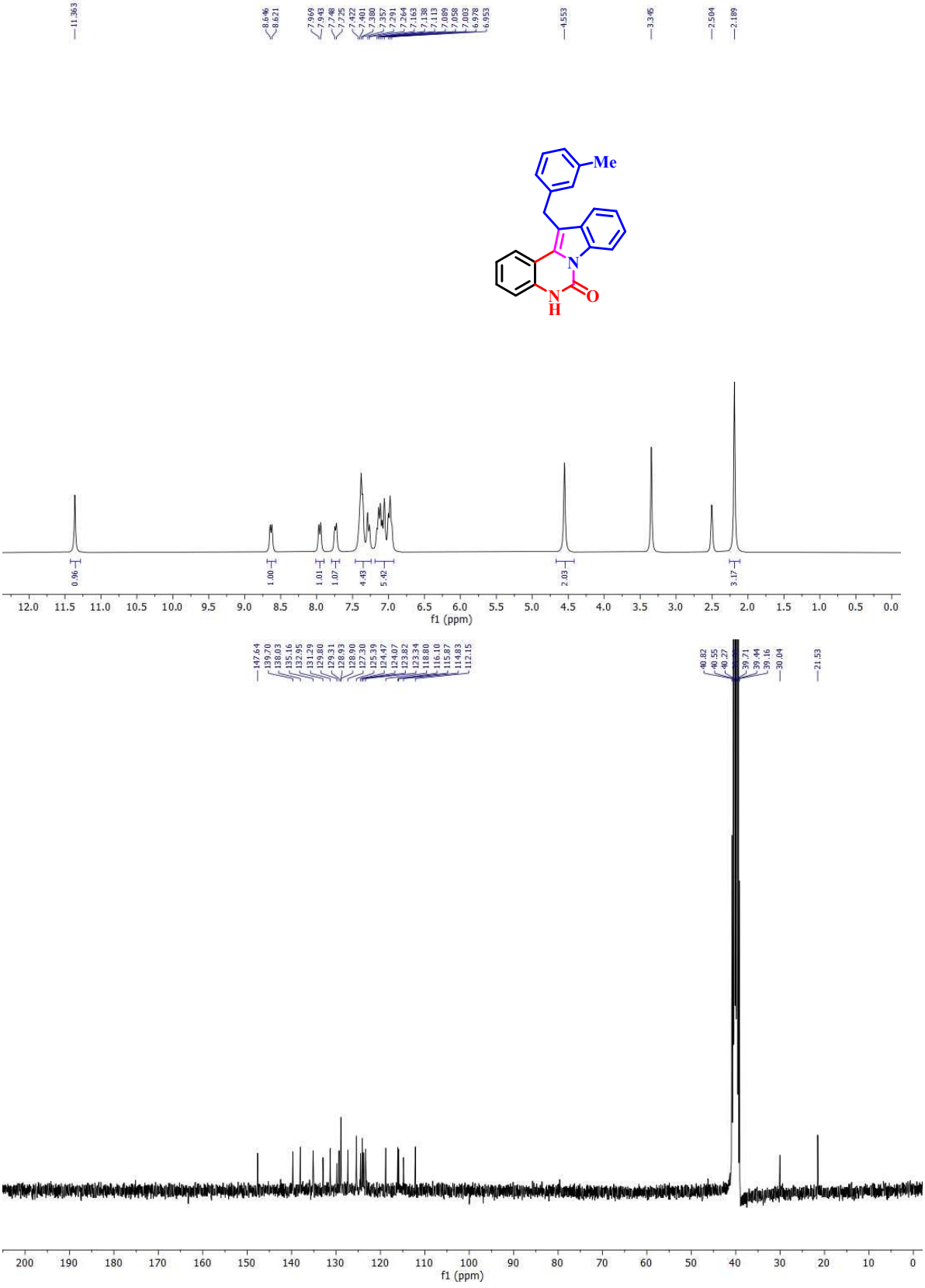
^{1}H and ^{13}C-NMR of compound (**4fe**)



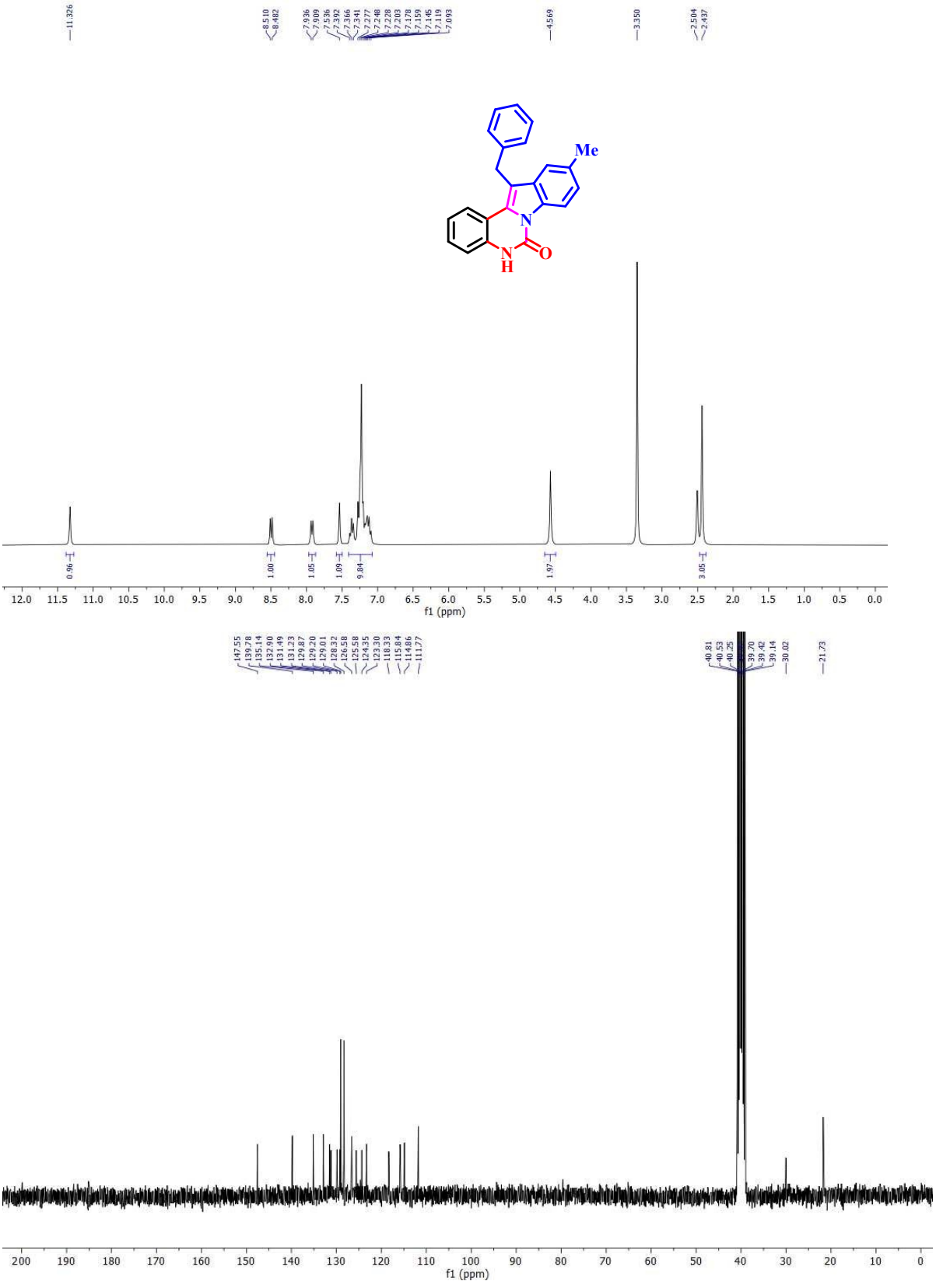
^{1}H and ^{13}C-NMR of compound (**5aa**)



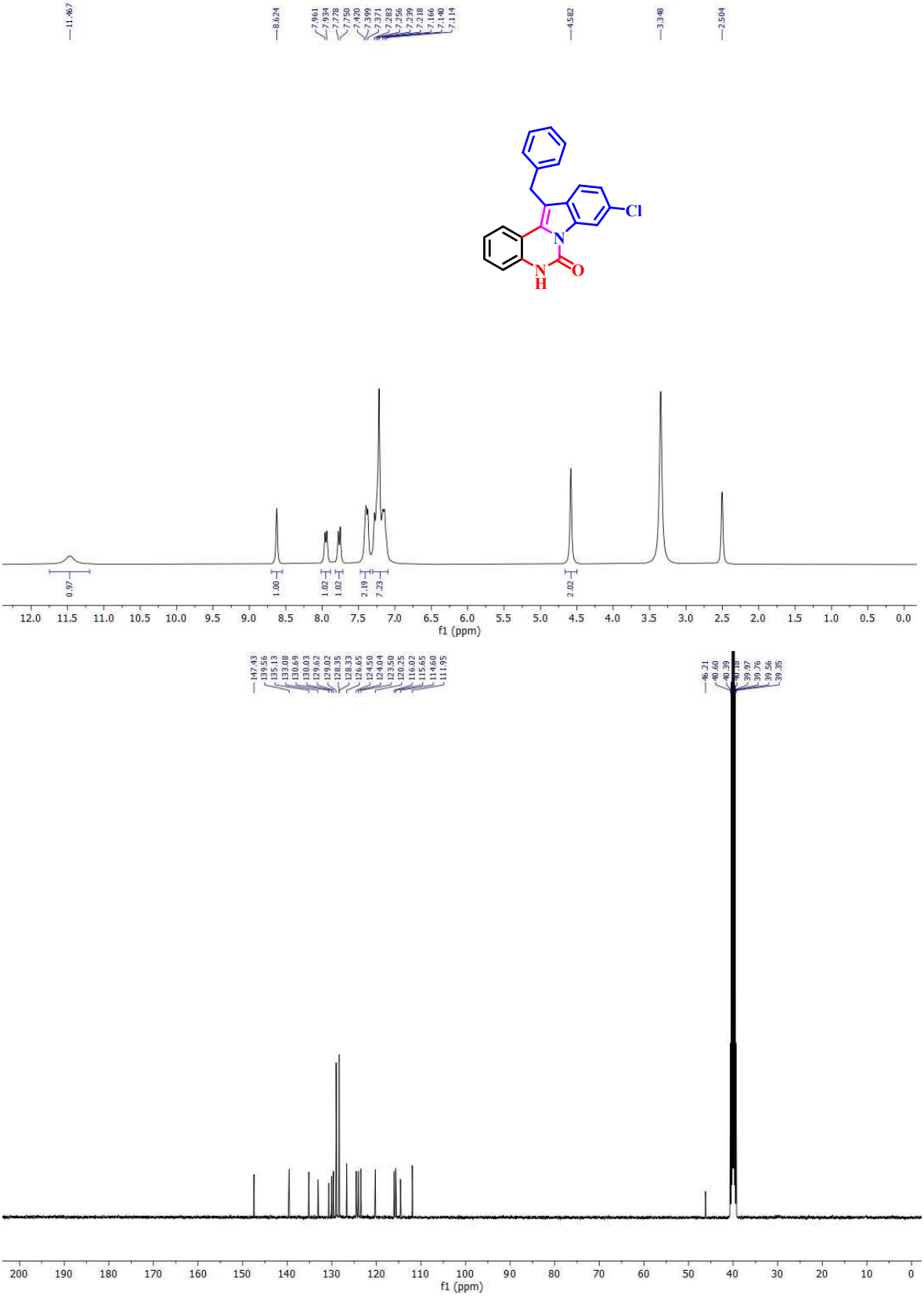
^{1}H and ^{13}C-NMR of compound (**5ah**)



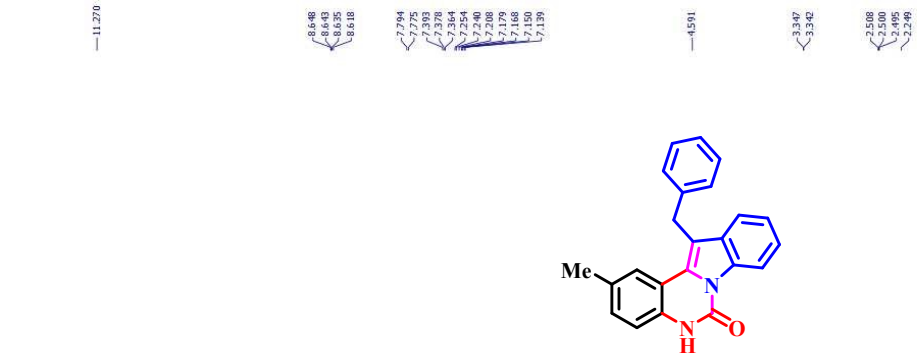
^{1}H and ^{13}C-NMR of compound (**5af**)

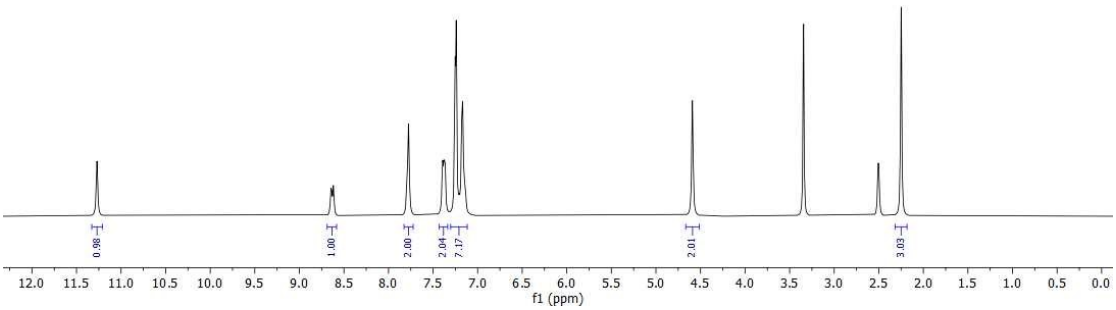


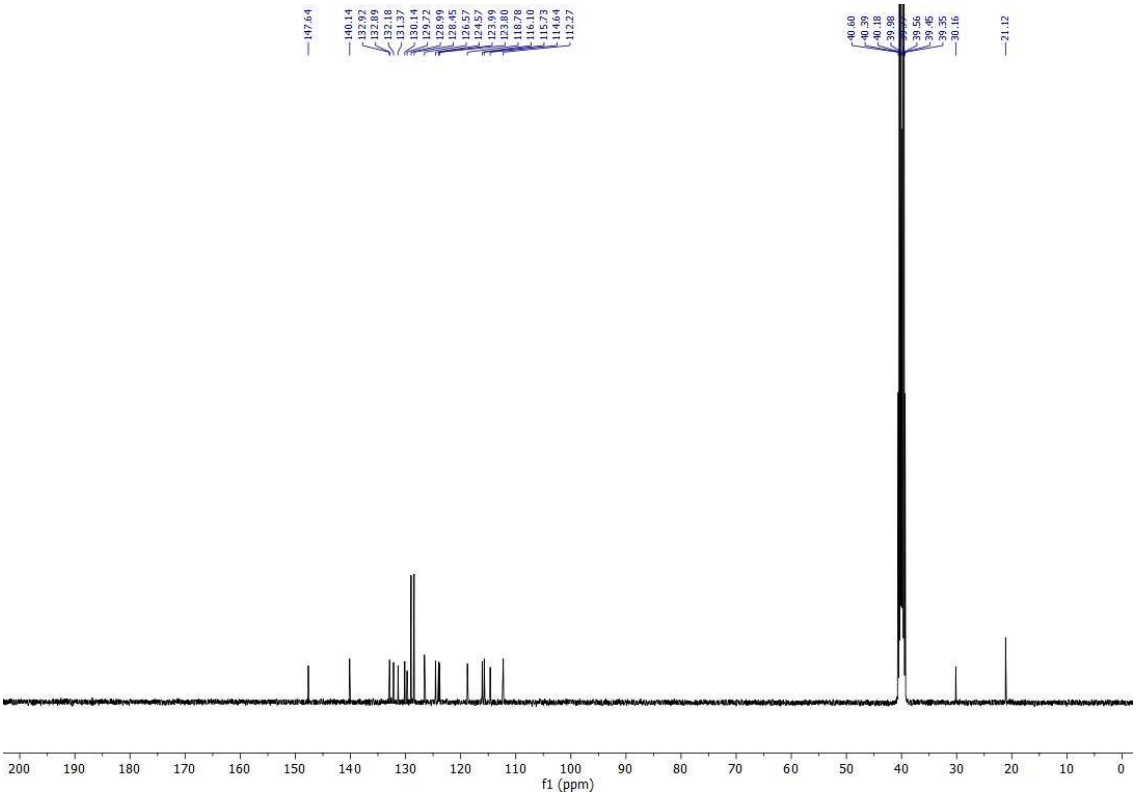
^{1}H and ^{13}C-NMR of compound (**5ag**)



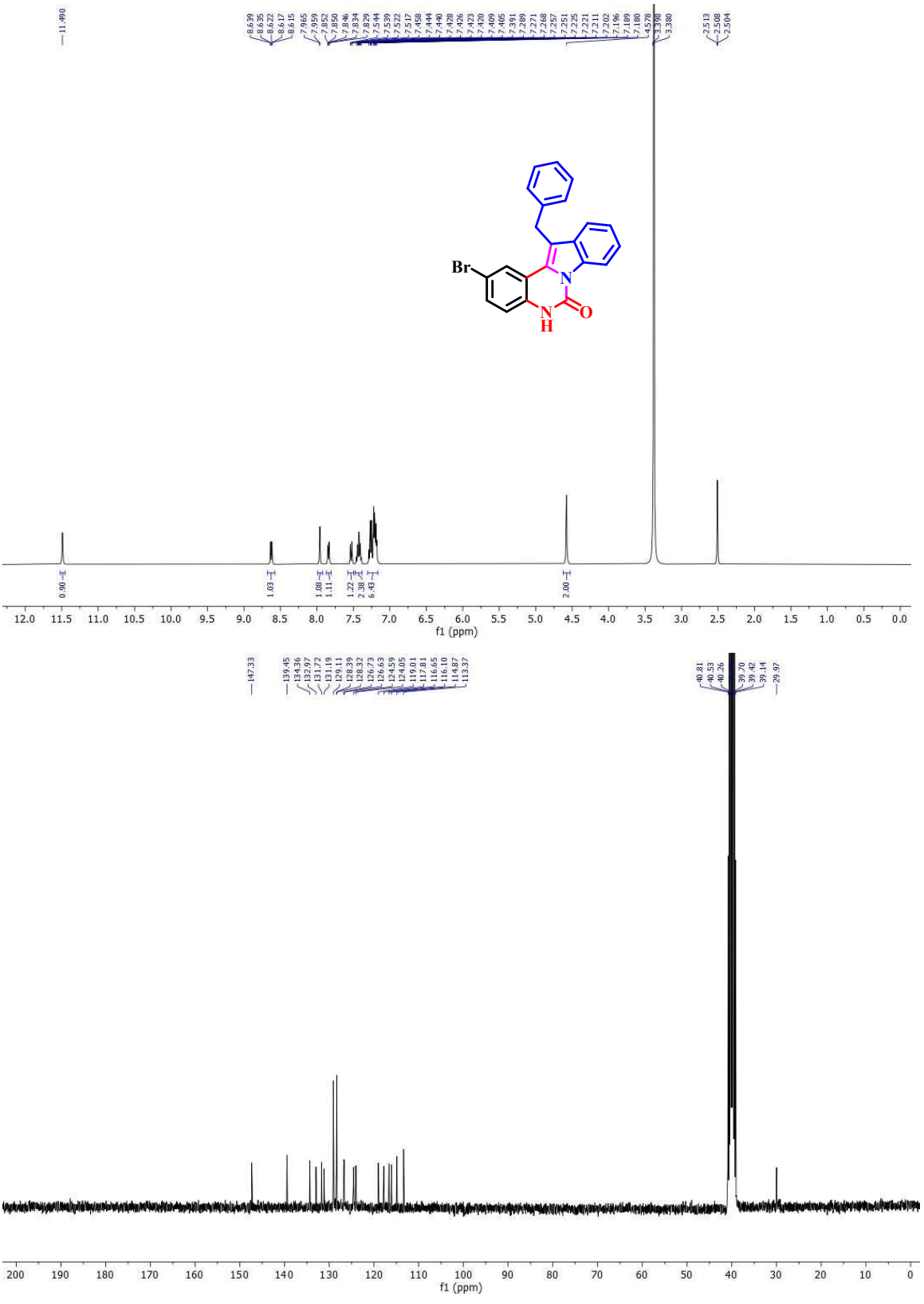
^{1}H and ^{13}C-NMR of compound (**5ba**)



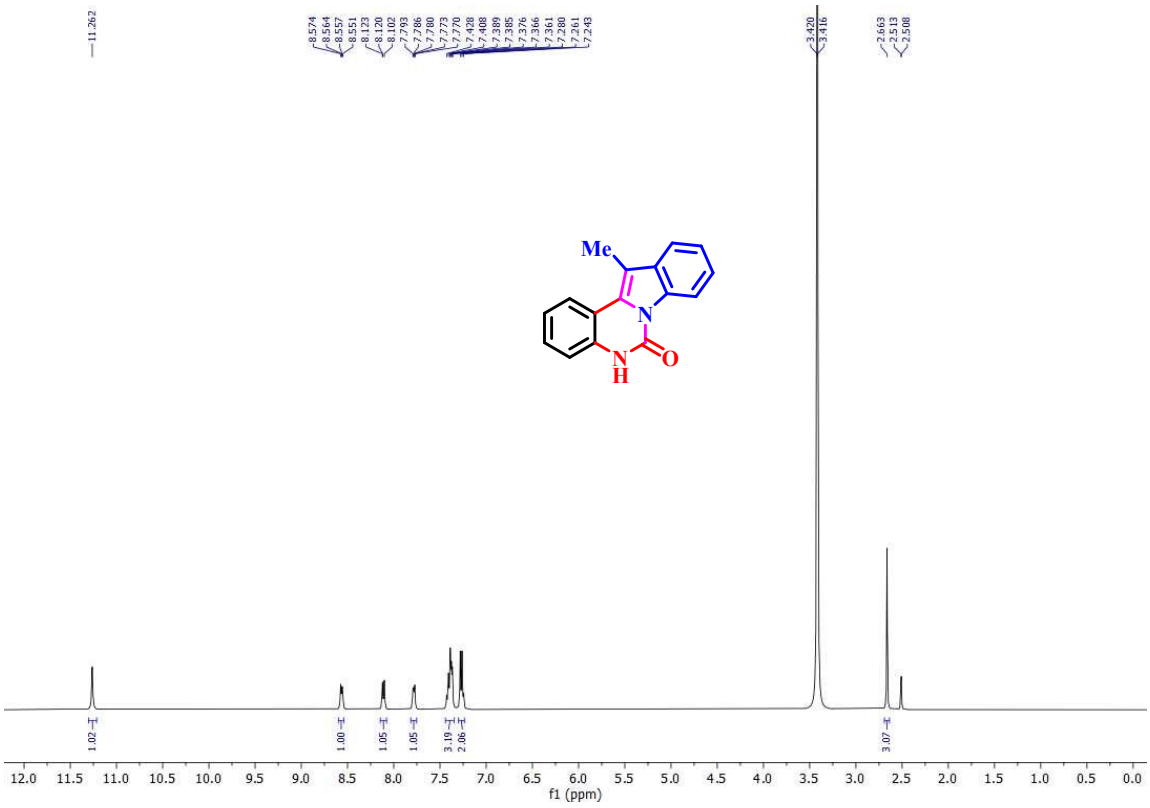


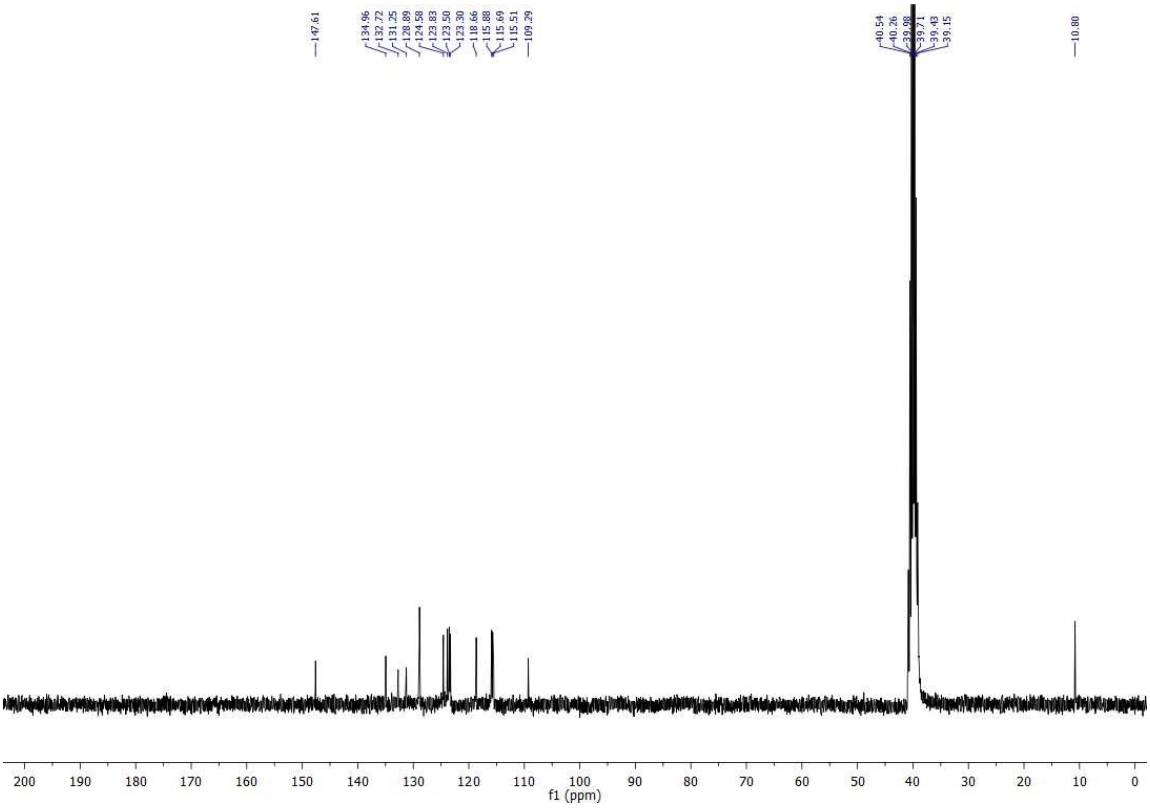


^{1}H and ^{13}C-NMR of compound (**5fa**)

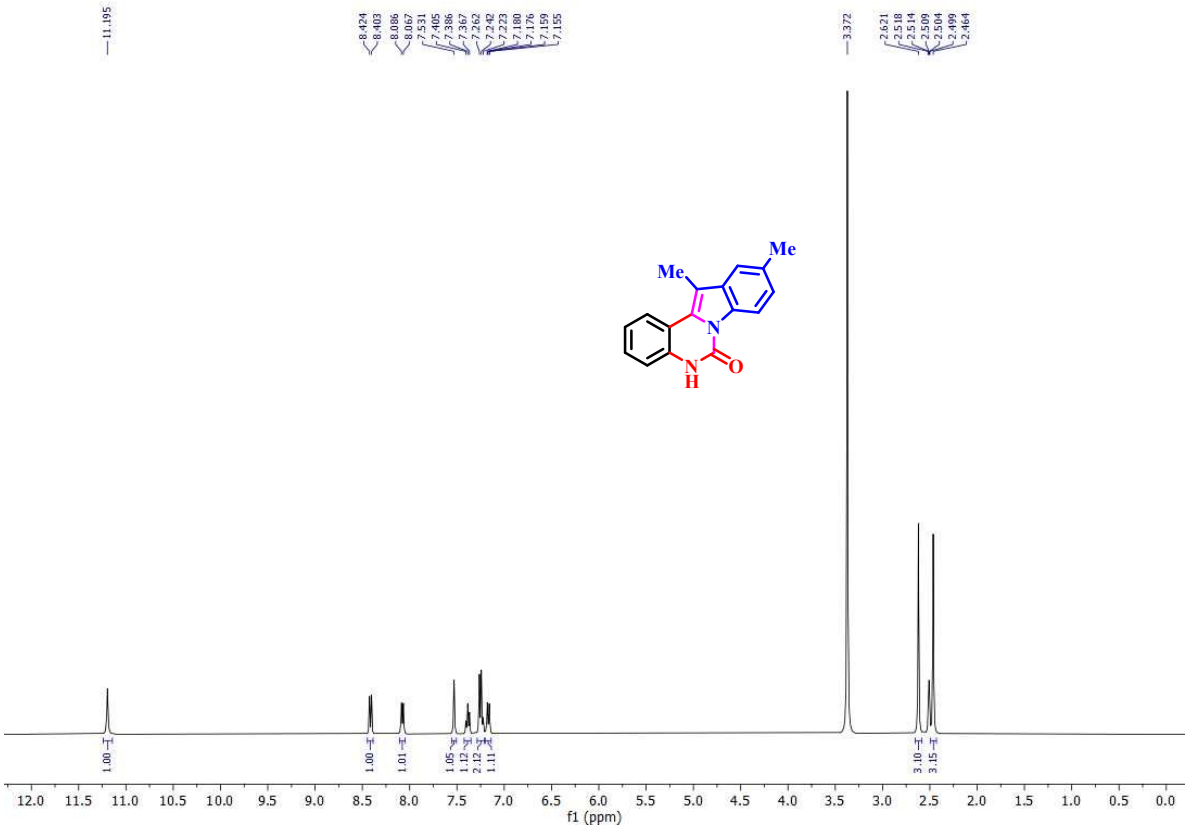


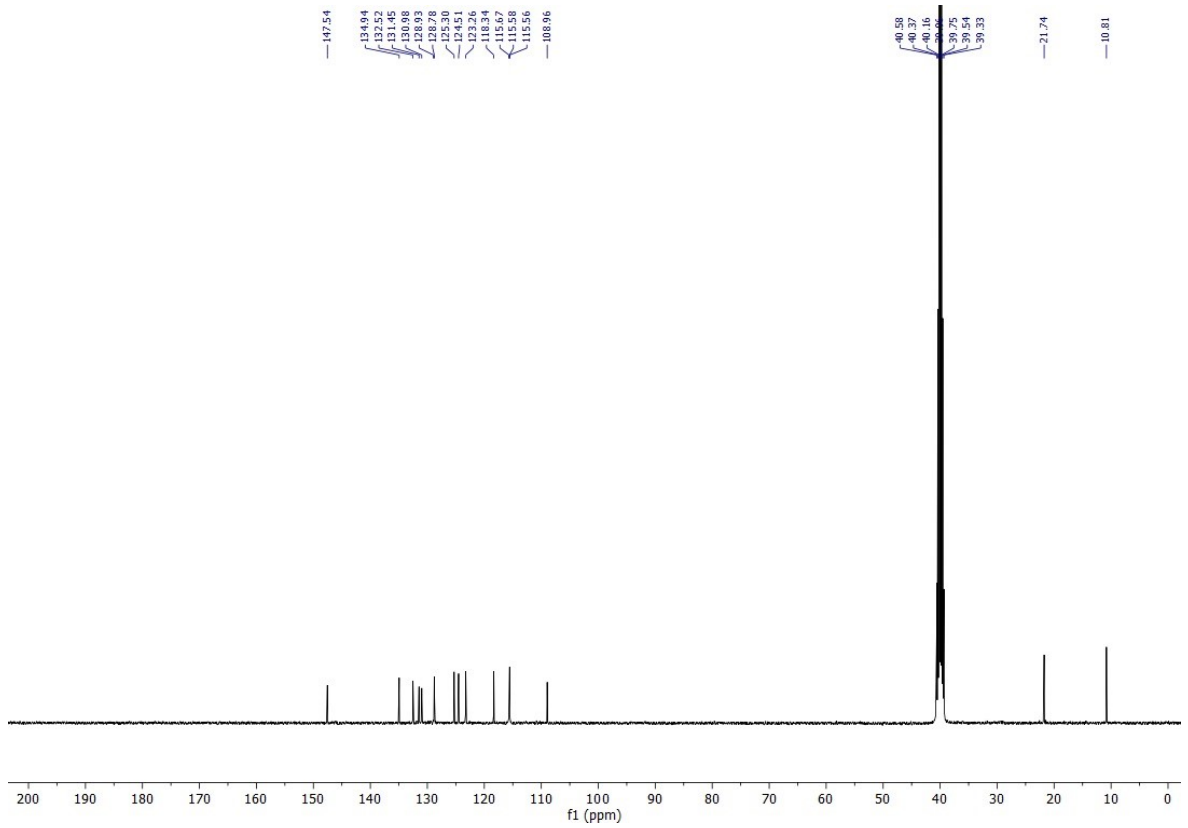
^{1}H and ^{13}C-NMR of compound (**6aa**)



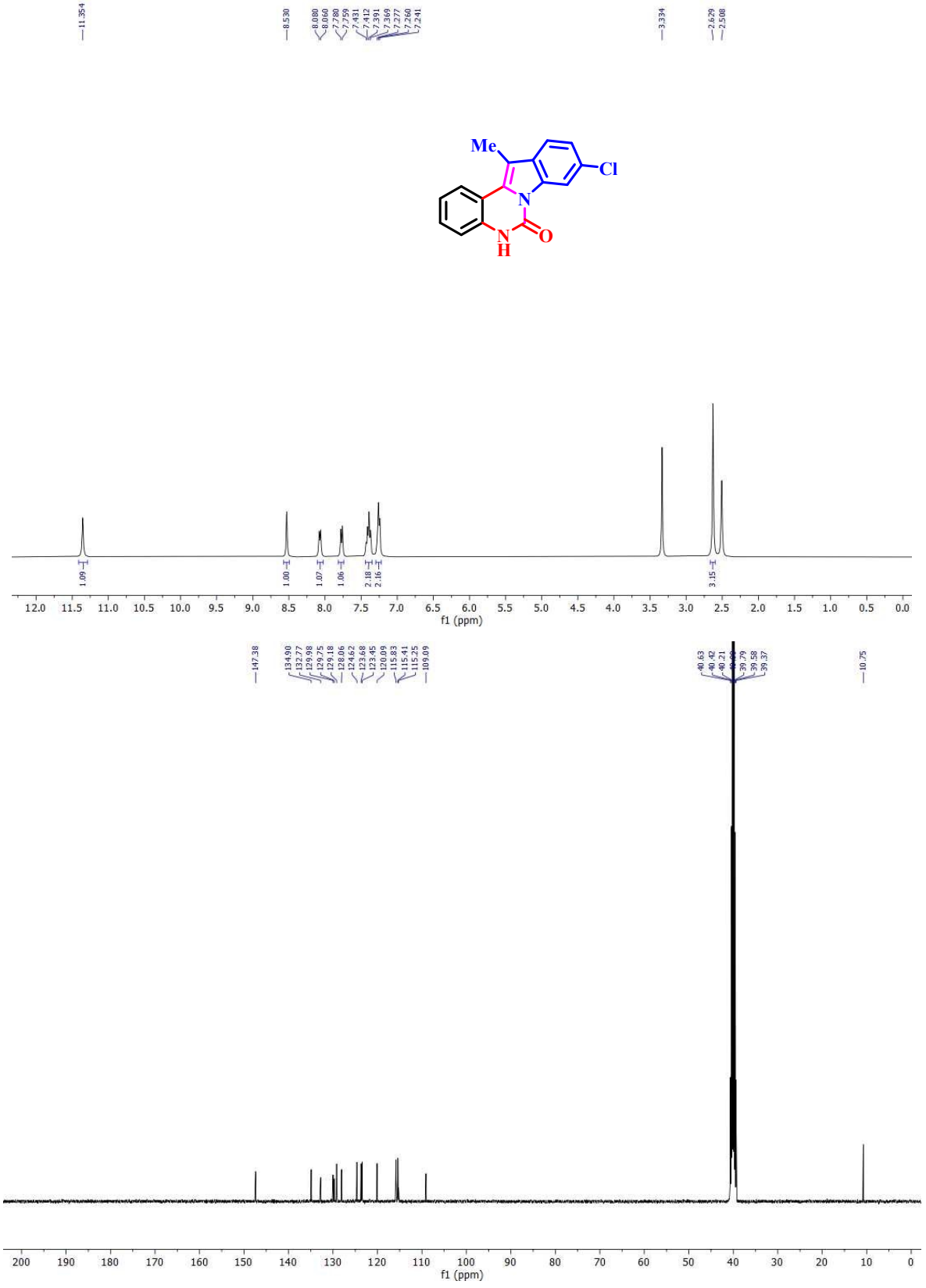


^{1}H and ^{13}C-NMR of compound (**6ab**)

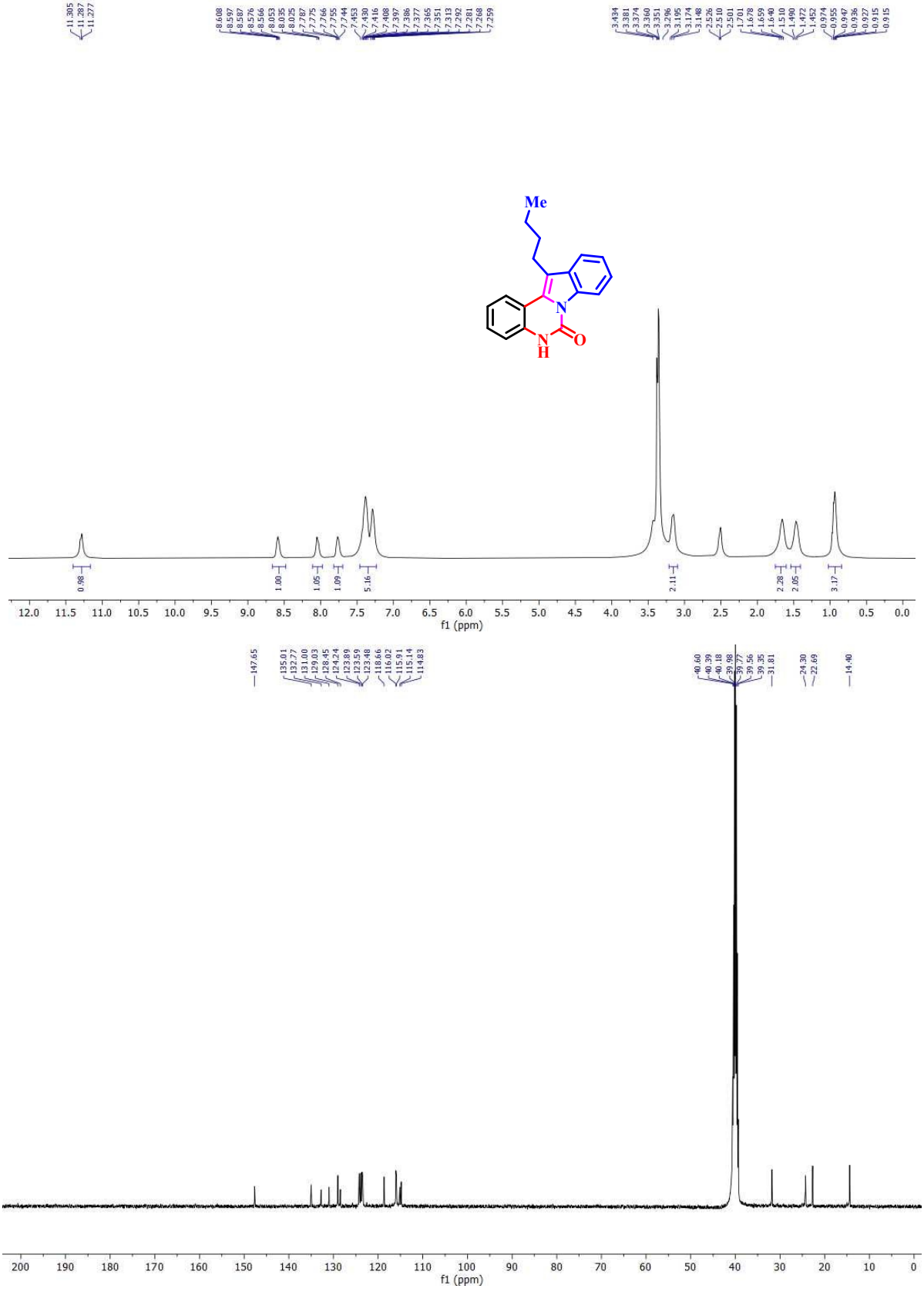




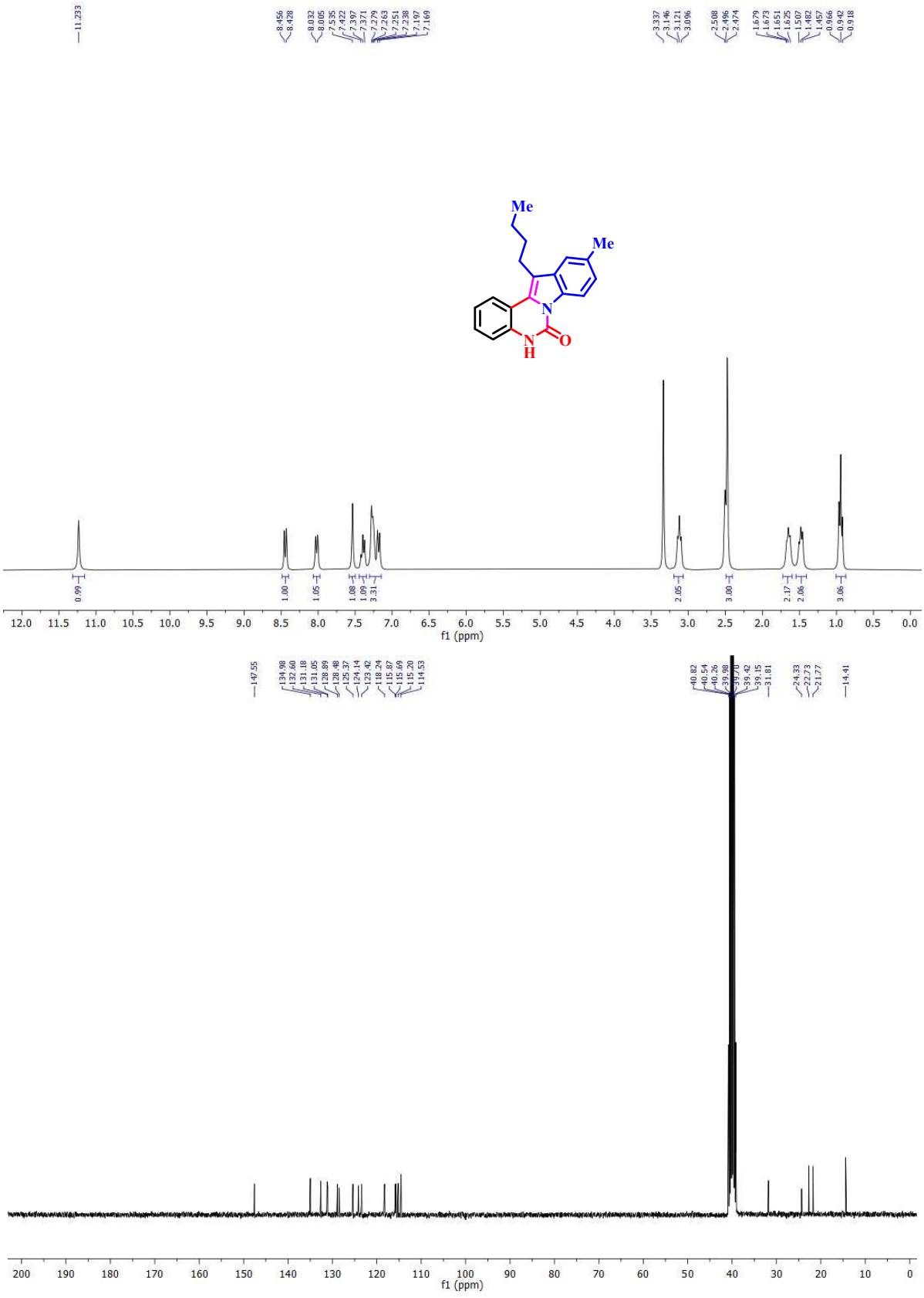
^{1}H and ^{13}C-NMR of compound (**6ac**)



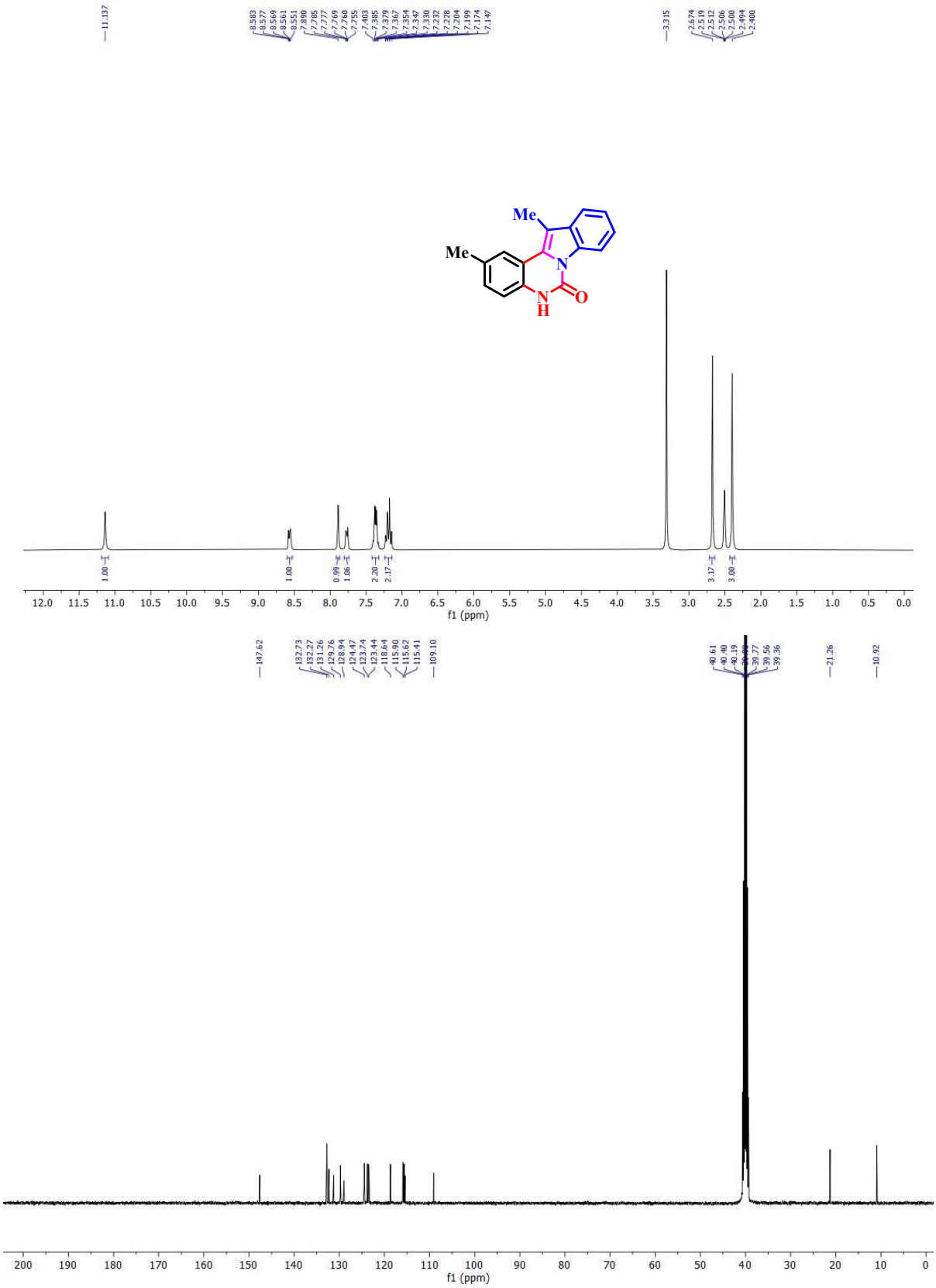
^{1}H and ^{13}C-NMR of compound (**6ad**)



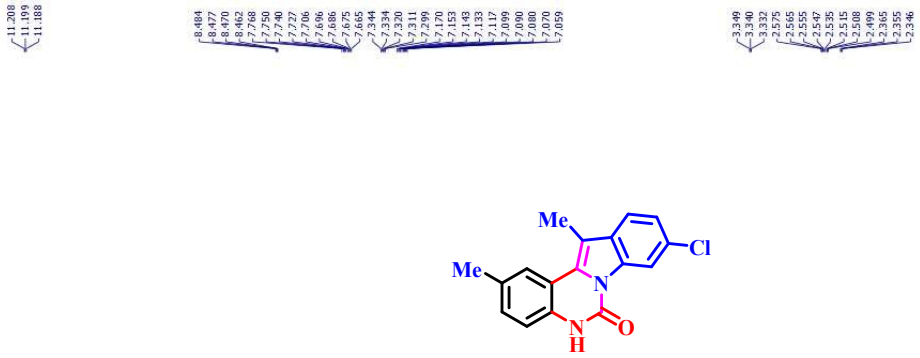
^{1}H and ^{13}C-NMR of compound (**6ae**)

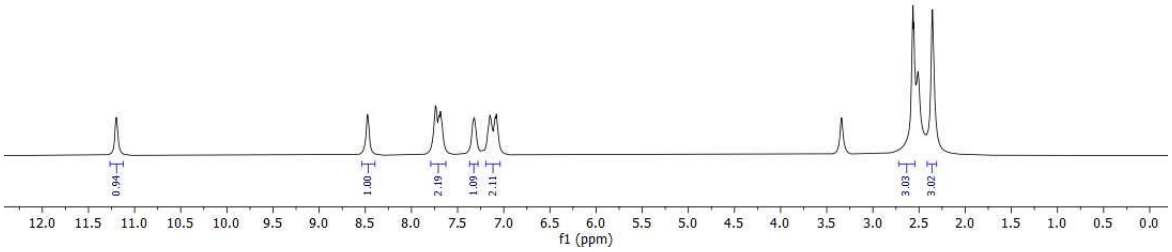


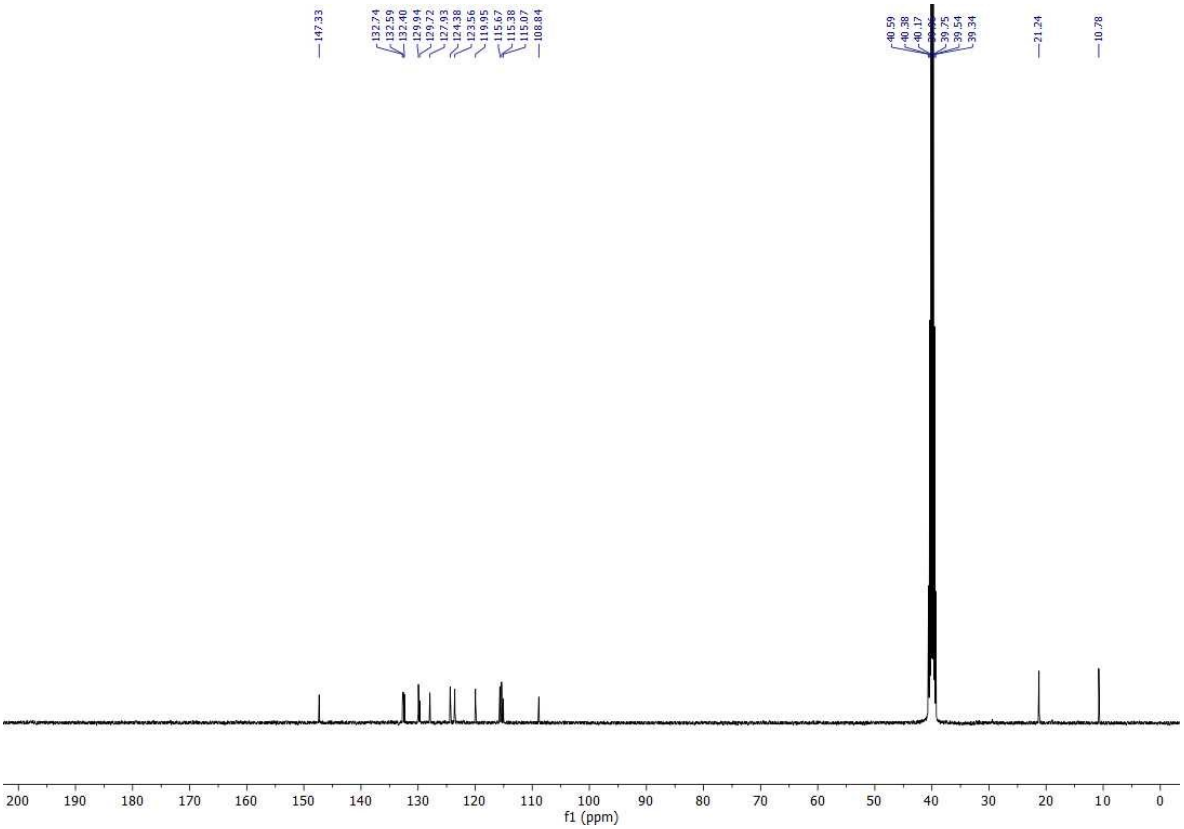
^{1}H and ^{13}C-NMR of compound (**6ba**)



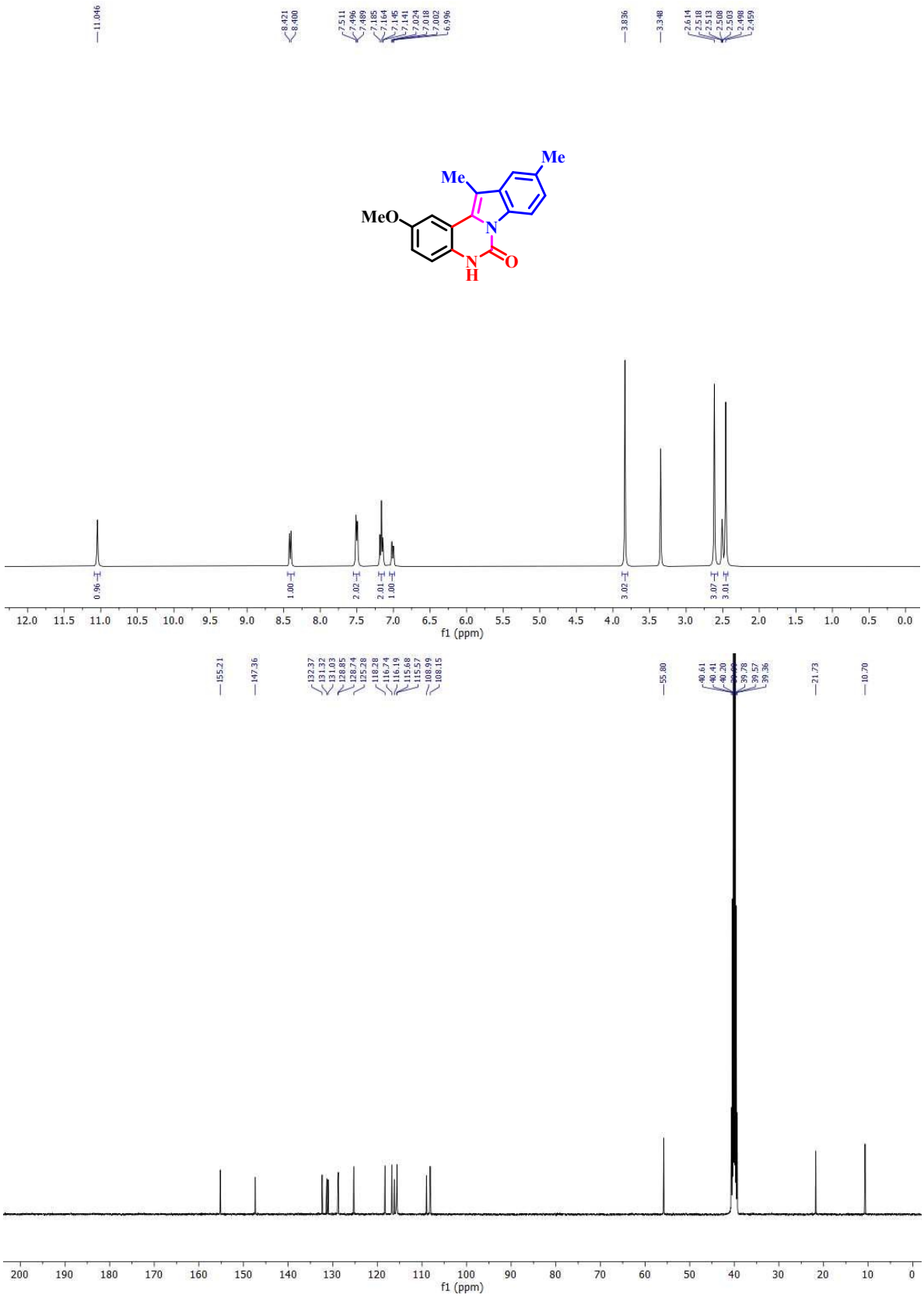
^{1}H and ^{13}C-NMR of compound (**6bc**)



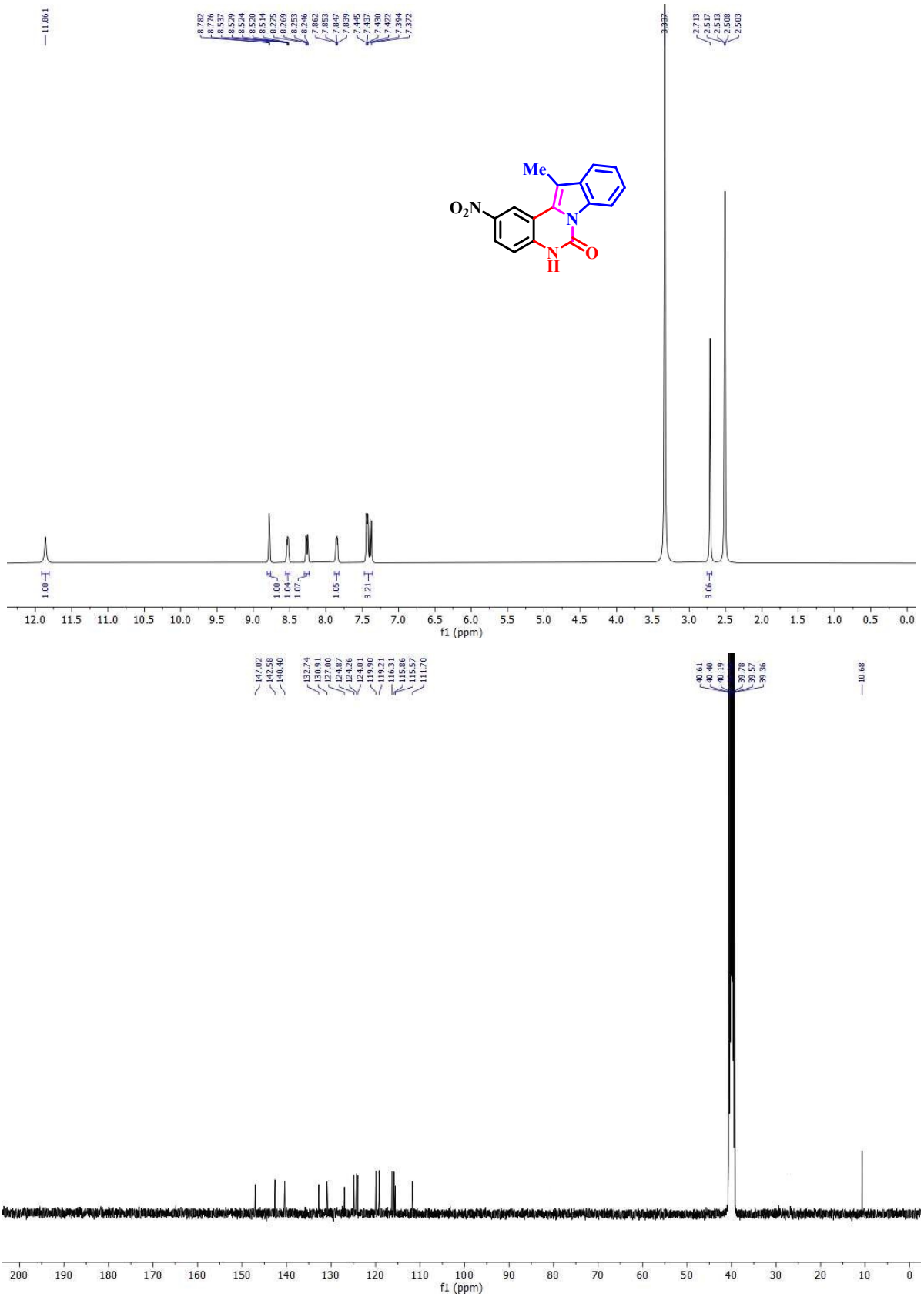




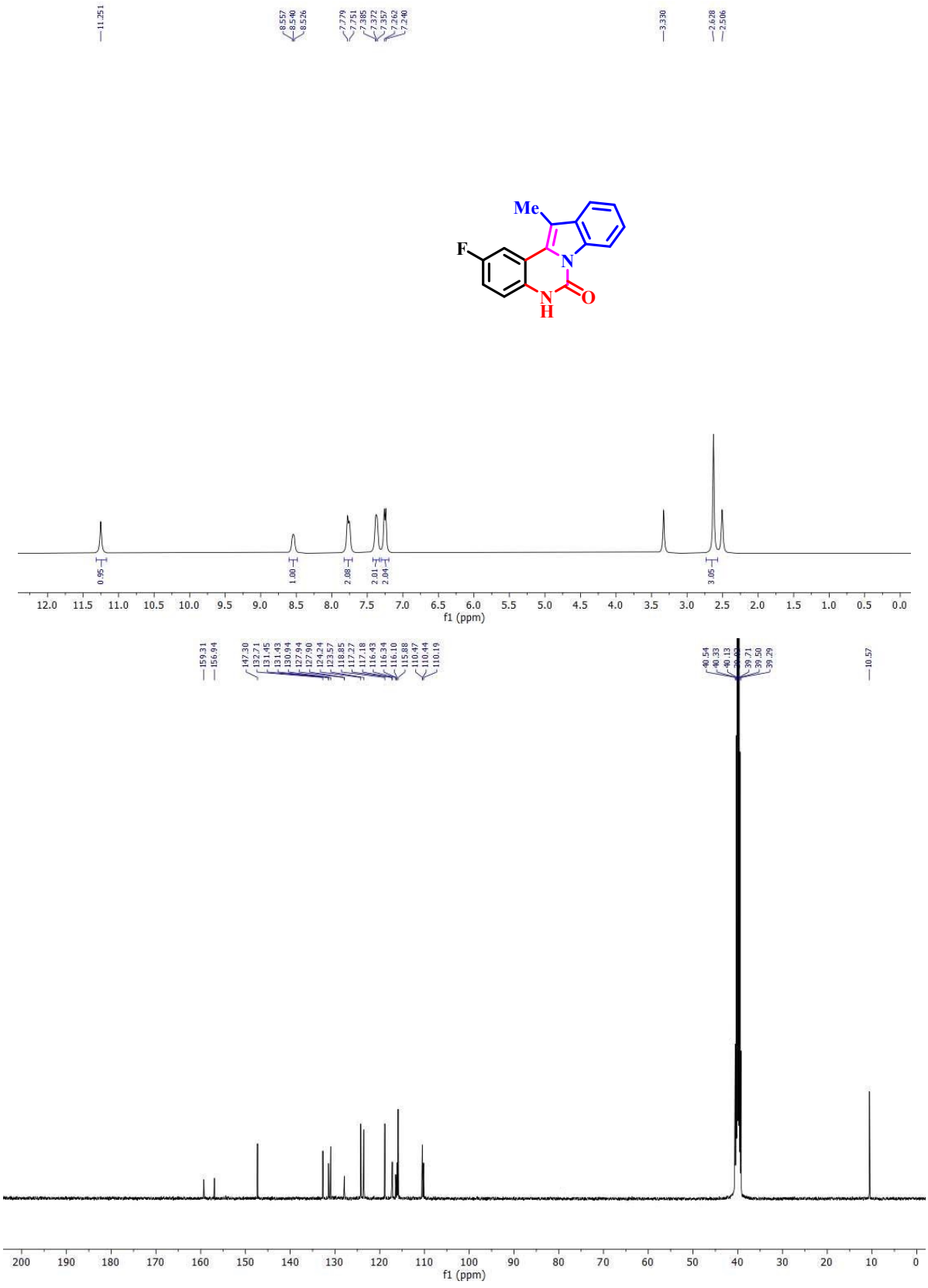
^{1}H and ^{13}C-NMR of compound (**6cb**)



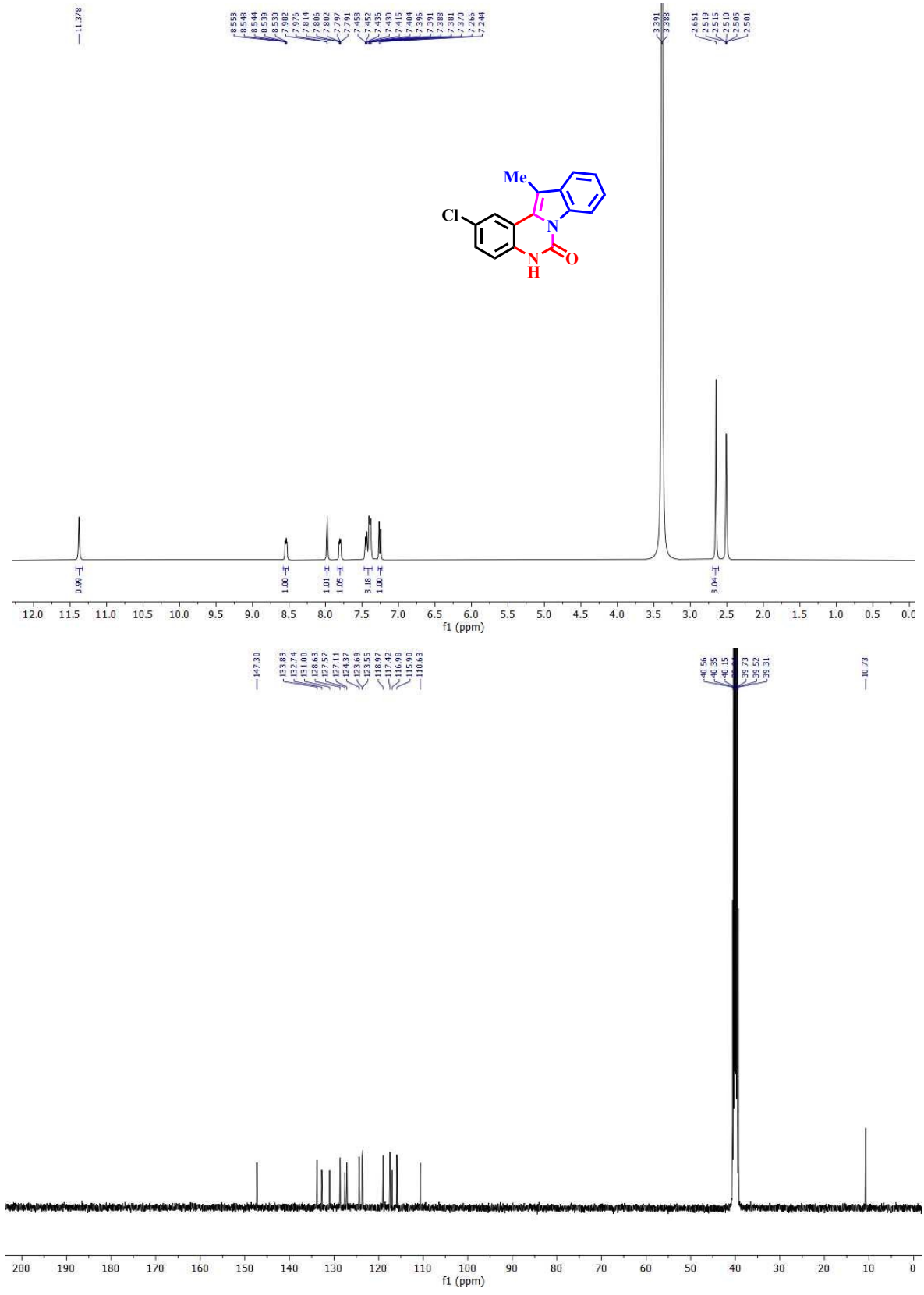
^{1}H and ^{13}C-NMR of compound (**6ga**)



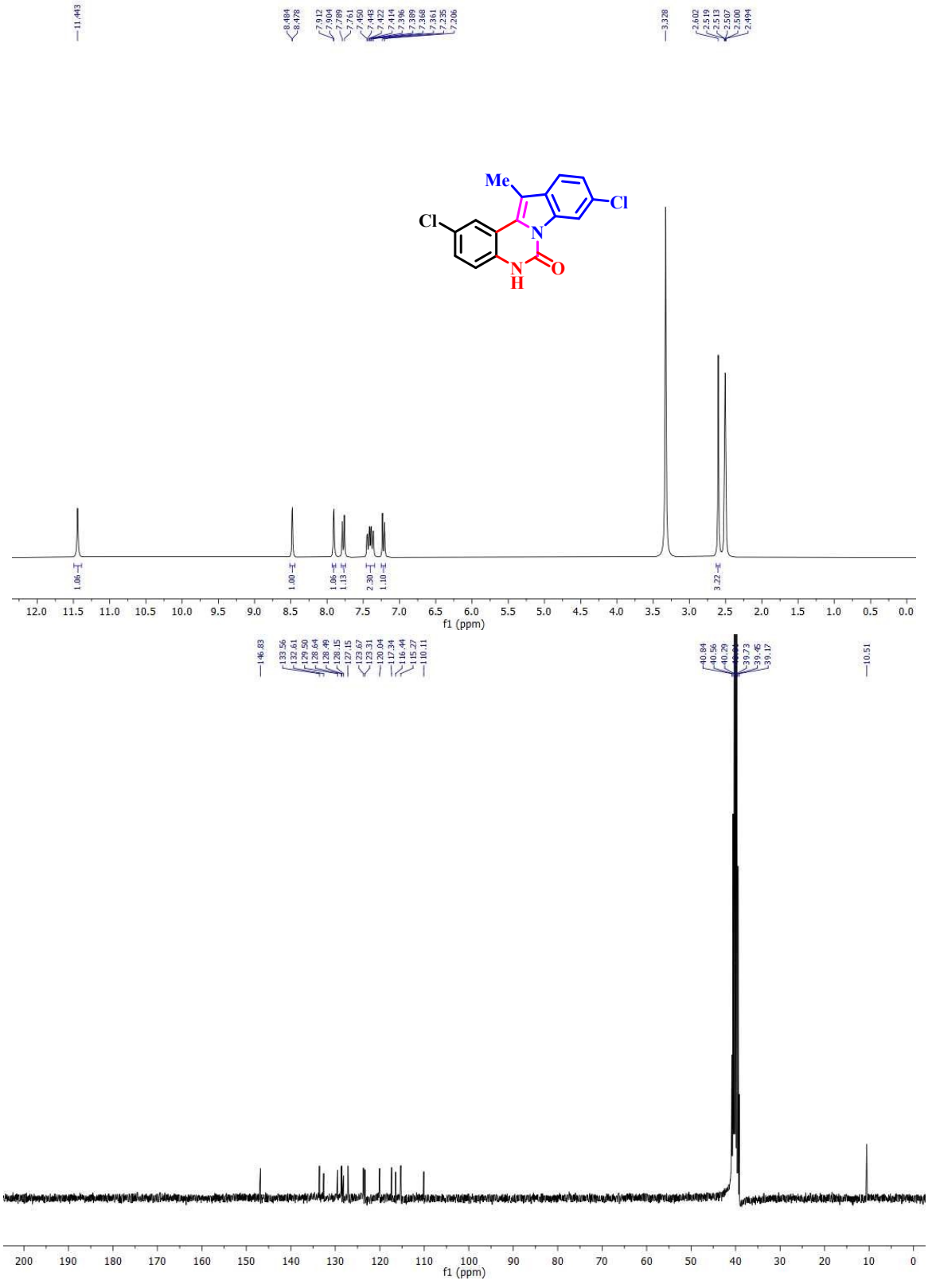
^{1}H and ^{13}C-NMR of compound (**6da**)



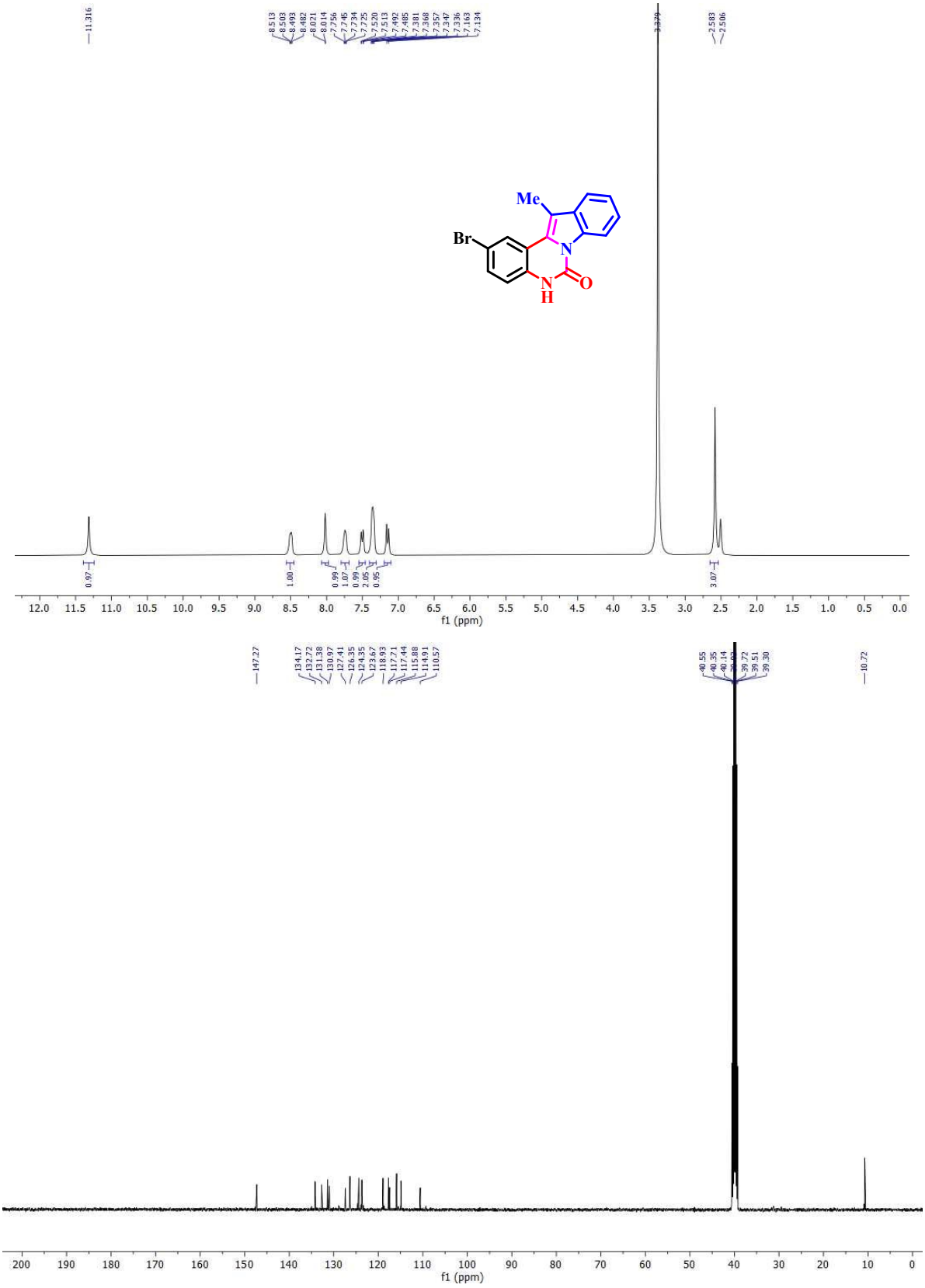
^{1}H and ^{13}C-NMR of compound (**6ea**)



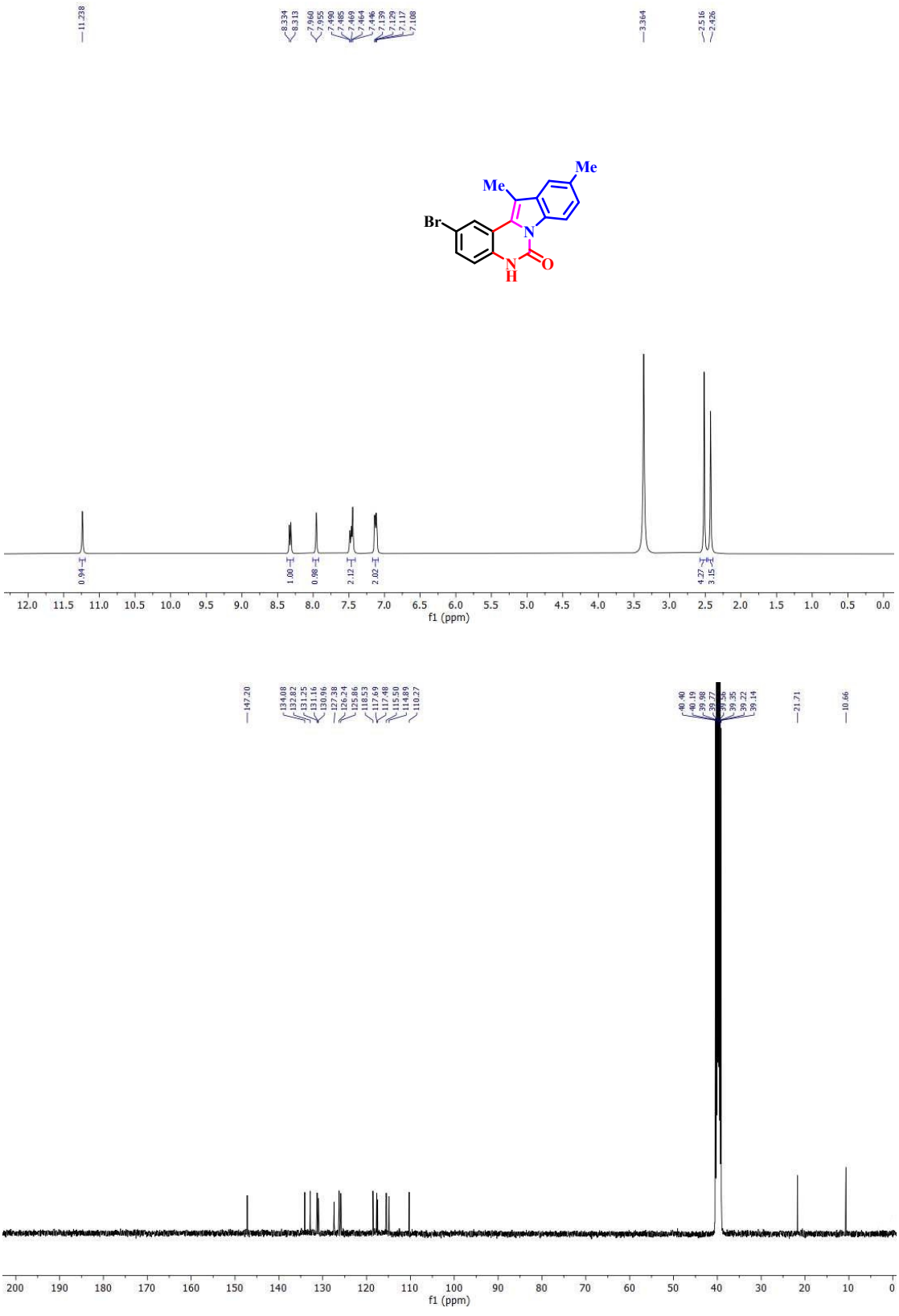
^{1}H and ^{13}C-NMR of compound (**6ec**)



^{1}H and ^{13}C-NMR of compound (**6fa**)

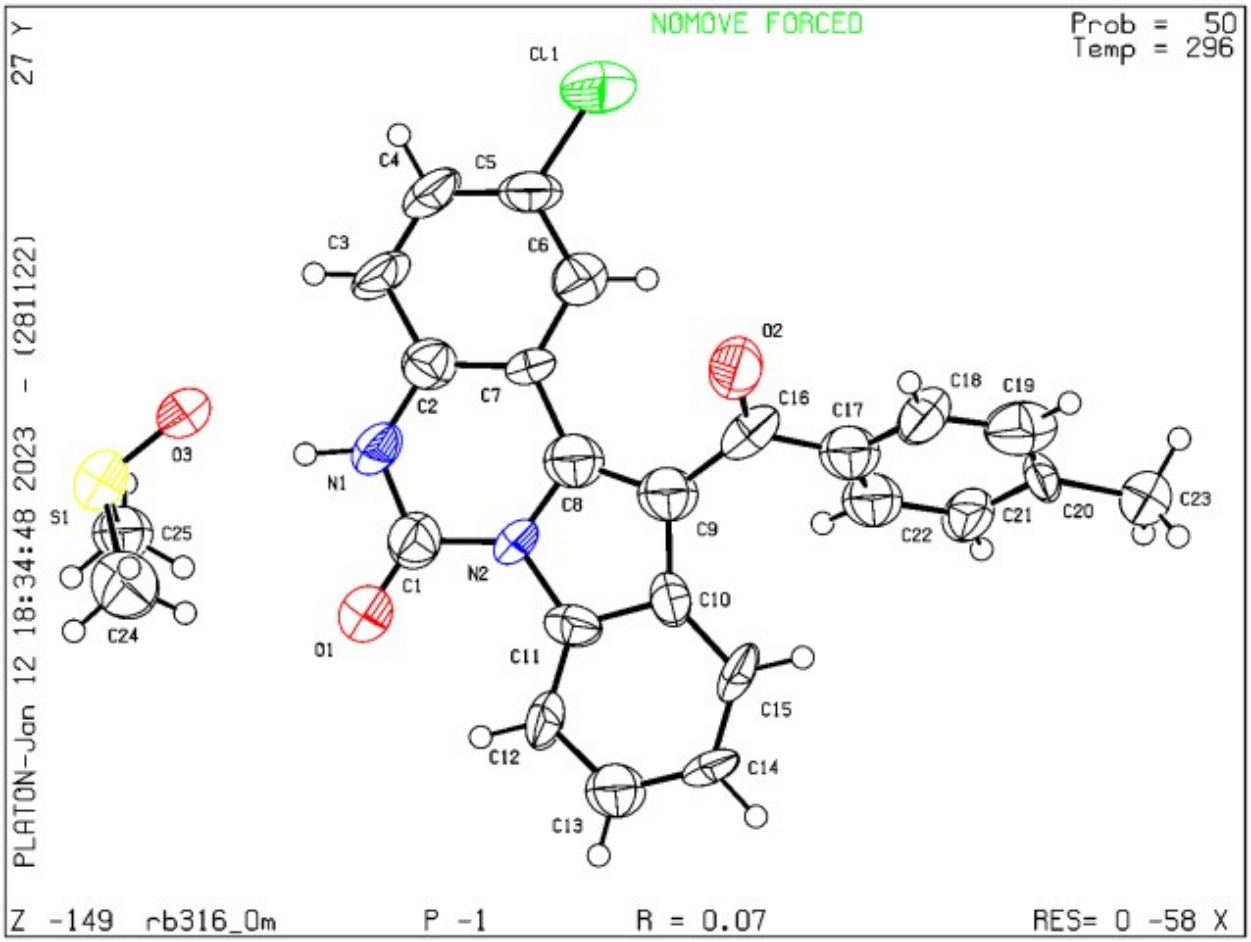


^{1}H and ^{13}C-NMR of compound (**6fb**)



11. X-ray Characterization Data Summery of Compound 4eb

Figure S2. ORTEP diagram of compound **4eb** (**CCDC 2240050**). Ellipsoids are drawn at 50% probability level.



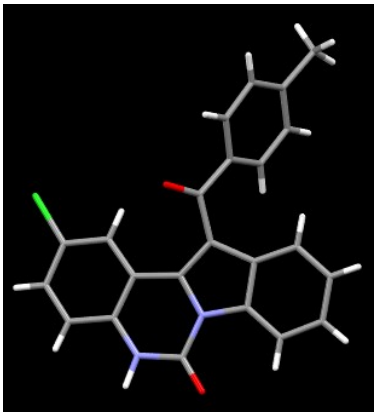


Table S1. Crystal Data and Structure Refinement for 4eb(CCDC 2240050).

Datablock: rb316_0m

Bond precision: C-C = 0.0242 A Wavelength=0.71073

Cell: a=10.012(6) b=10.023(8) c=11.978(6)
alpha=94.38(6) beta=109.83(6) gamma=93.94(6)
Temperature: 296 K

	Calculated	Reported
Volume	1121.7(13)	1121.6(13)
Space group	P -1	P -1
Hall group	-P 1	-P 1
Moiety formula	C23 H15 Cl N2 O2, C2 H6 O S	C23 H15 Cl N2 O2, C2 H6 O S
Sum formula	C25 H21 Cl N2 O3 S	C25 H21 Cl N2 O3 S
Mr	464.95	464.95
Dx,g cm-3	1.377	1.377
Z	2	2
Mu (mm-1)	0.294	0.294
F000	484.0	484.0
F000'	484.73	
h,k,lmax	8,8,9	7,7,9
Nref	1223	1182
Tmin,Tmax	0.929,0.935	0.929,0.935
Tmin'	0.929	

Correction method= # Reported T Limits: Tmin=0.929 Tmax=0.935
AbsCorr = MULTI-SCAN

Data completeness= 0.966 Theta(max)= 16.644

R(reflections)= 0.0720(760) wR2(reflections)= 0.2164(1182)

S = 1.005 Npar= 262

Crystal Structure Experimental Protocol:

Single crystal of compound **4eb** was mounted on a Bruker-AXS SMART APEX II diffractometer equipped with a graphite monochromator and Mo Kα (λ = 0.71073 Å) radiation. The crystal was placed 60 mm from the CCDC, and frames (360) were measured with a counting time of 6s. The structures were solved using the Patterson method using the SHELXS 97 program. Non-hydrogen atoms were refined with independent anisotropic displacement parameters, while difference Fourier synthesis and least-squares refinement showed the positions of any remaining non-hydrogen atoms. The non-hydrogen atoms

were refined with anisotropic thermal parameters. Successful convergence was indicated by the maximum shift/error of 0.001 for the last cycle of the least-squares refinement. All other crystallographic details such as h, k, l ranges, 2θ ranges, and R-factors can be found above (Table S1).

1.Reference

(1)Wang,X.; Li, J.; Huang,Y.; Zhu,J.; Hu, R.; Wu,W.; Jiang,H.Facile Synthesis of π-Conjugated Quinazoline-Substituted Ethenes from 2-Ethynylanilines and Benzonitriles under Transition-Metal-Free Conditions. *J. Org. Chem*. **2018**, *83*, 10453.