Supporting information

For

Cascade [4+1] annulation via greener nitrogen ylide in water: Synthesis of bicyclic and tricyclic fused dihydrofuran

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Contents

General remarks

Commercially available *N*-methylimidazole from Aldrich was used. Progress of reactions was monitored by thin layer chromatography (TLC). NMR spectra were recorded in d_6-DMSO or $CDCl_3$ at 300 and 200 MHz (based on availability of instruments) 75 and 50 MHz (for ^{13}C) respectively on Bruker Avance DPX-300 MHz and Bruker Avance DPX-200 MHz. Chemical shifts are reported in δ (ppm) relative to TMS (^1H) or $CDCl_3$ (^{13}C) as internal standards. Integrals are in accordance with assignments; coupling constants are given in Hz. Yields refer to quantities obtained after chromatography.

General experimental procedure

Typical procedure for the preparation of 1-methyl-3-phenacylimidazolium bromide (3):

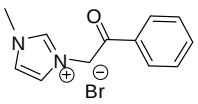
A solution of 8.2 g (0.1 mol) of *N*-methylimidazole and 20.0 g (0.1 mol) of phenacylbromide in 300 ml of ether was allowed to stand at room temperature for 16h. The solids, which separated, was collected and then recrystallized from acetonitrile to give 27.0 g (95%) of 1-Methyl-3-phenacylimidazolium bromide as white powder.

Representative one pot procedure for the synthesis of 6a-h, 8a-d, 10a-j:

A mixture of substituted aldehyde (1 mmol), 4-hydroxy coumarin / 5, 5 dimethyl, 1, 3-cyclohexanedione / 1, 3-cyclohexanedione / 4-hydroxy-6-methyl-2*H*-pyran-2-one (1 mmol) and *N*-methyl imidazole (0.5 mmol) in water (5 ml) was allowed to reflux for 1h at 100ºC. After that 1-methyl-3-phenacylimidazolium bromide (1 mmol) was added and stirring was continued for 1h at 100ºC. After completion of reaction as indicated on TLC, the reaction mixture was extracted with ethyl acetate and water. Organic layer was dried over anhydrous sodium sulphate and concentrated in vacuo. The crude product was chromatographed on a silica gel column with a hexane-ethyl acetate mixture to afford dihydrofuran derivatives **6a-h, 8a-d,** and **10a-j** in good to excellent yield.

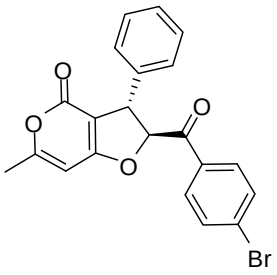
Characterisation data of all the compounds

***N*-Methyl-3-phenacylimidazolium bromide (3):**



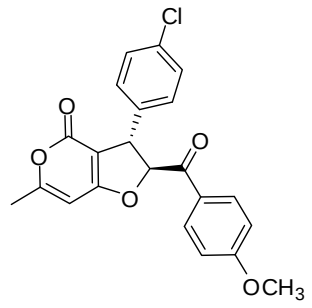
m.p:153-155°C; 95% as white solid; ^{1}H NMR (DMSO-d_6, 300 MHz) δ = 9.11 (s, 1H, CH), 8.07 (d, *J* = 7.3Hz, 2H, ArH), 7.80-7.73 (m, 3H, Ar-H), 7.66 (t, *J* = 7.7Hz, 2H, CH), 6.10 (s, 2H, CH_2), 3.96 (s, 3H, N-CH_3); ^{13}C NMR (DMSO-d_6, 50 MHz) δ = 191.4, 137.7, 134.5, 133.7, 129.1, 128.2, 123.9, 123.3, 55.4, 36.0; Ana. Calcd for $C_{12}H_{13}N_2OBr$: C, 51.26; H, 4.66; N, 9.96. Found: C, 51.11; H, 4.58; N, 10.02.

2-(4-Bromobenzoyl)-6-methyl-3-phenyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6a): m.p: 152-155°C; White solid; yield: 95% ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.72 (d, *J* = 8.9Hz, 2H, Ar-H), 7.63 (d, *J* = 7.1Hz, 2H, ArH), 7.37 (d, *J* = 7.3Hz, 3H, ArH), 7.25 (d, *J* = 8.8Hz, 2H, ArH), 6.14 (s 1H, CH), 5.93 (d, *J* = 4.8Hz, 1H, CH), 4.63 (d, *J* = 4.6Hz, 1H, CH), 2.29 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 191.5, 170.9, 166.8, 160.7, 139.7, 132.4, 130.5, 129.3, 128.2, 127.5, 102.5, 95.5, 92.2, 48.3, 20.6; MS (ESI+) *m/z*: 411.0 $(M+H)^+$ Ana. Calcd for $C_{21}H_{15}BrO_4$: C, 61.33; H, 3.68; Found: C, 61.38; H, 3.60.



2-Benzoyl-3-(4-fluorophenyl)-6-methyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6b): m.p:135-140°C; White solid; yield: 86%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.85 (d, *J* = 7.1Hz, 2H, Ar-H), 7.64 (d, *J* = 6.5Hz, 1H, ArH), 7.51 (d, *J* = 7.4Hz, 2H, ArH), 7.24 (dd, *J* = 5.4Hz, 2H, ArH), 7.08 (m, 2H, ArH), 6.16 (s, 1H, CH), 5.96 (d, *J* = 4.9Hz, 1H, CH), 4.63 (d, *J* = 4.7Hz, 1H, CH), 2.30 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 192.3, 171.1, 166.9, 164.9, 160.8, 160.0, 135.6, 134.5, 133.2, 129.3, 129.1, 116.4, 116.0, 102.4, 95.6, 92.3, 47.7, 20.6; MS (ESI+) *m/z*: 351.1 (M+H)+ Ana. Calcd for $C_{21}H_{15}FO_4$: C, 71.99; H, 4.32; Found: C, 71.91; H, 4.38.

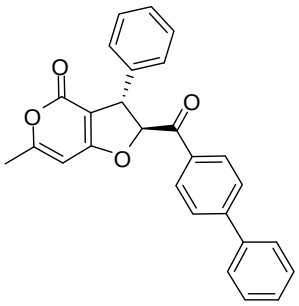
3-(4-Chlorophenyl)-2-(4-methoxybenzoyl)-6-methyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one



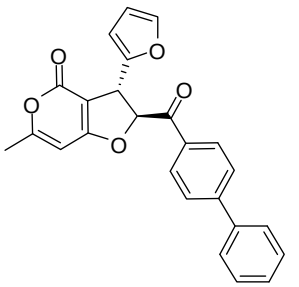
(6c): m.p:147-150°C; white solid; yield: 82%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.82 (d, *J* = 8.6Hz, 2H, Ar-H), 7.35 (d, *J* = 8.4Hz, 2H, ArH), 7.20 (d, *J* = 8.4Hz, 2H, ArH), 6.95 (d, *J* = 8.9Hz, 2H, Ar-H), 6.15 (s, 1H, CH), 5.90 (d, *J* = 5.0Hz, 1H, CH), 4.62 (d, *J* = 4.8Hz, 1H, CH), 3.88 (s, 3H, OCH_3), 2.29 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 190.6, 171.2, 166.9, 164.6, 160.8, 138.5,

133.9, 131.5, 129.4, 129.0, 126.0, 114.8, 102.2, 95.6, 91.9, 55.7, 48.0, 20.7; MS (ESI+) *m/z*: 397.1 $(M+H)^+$. Ana. Calcd for $C_{22}H_{17}ClO_5$: C, 66.59; H, 4.32; Found: C, 66.54; H, 4.28.

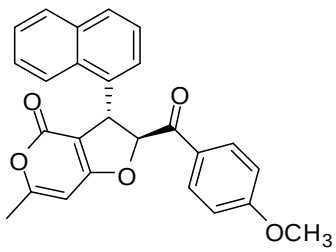
2-(Biphenylcarbonyl)-6-methyl-3-phenyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6d): m.p:155-160°C; white solid; yield: 91%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.85 (d, *J* = 8.3Hz, 2H, Ar-H), 7.64 (d, *J* = 8.2Hz, 2H, ArH), 7.56 (d, *J* = 6.9Hz, 2H, ArH), 7.43-7.34 (m, 3H, ArH), 7.29 (s, 2H, Ar-H), 7.18 (t, *J* = 7.9Hz, 3H, ArH), 6.09 (s, 1H, CH), 5.90 (d, J = 5.0Hz, 1H, CH), 4.60 (d, *J* = 4.8Hz, 1H, CH), 2.23 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ =191.8, 171.2, 167.0, 160.8, 147.2, 139.4, 138.4, 134.0, 131.8, 129.7, 129.5, 129.2, 129.0, 128.8, 127.7, 127.4, 102.3, 95.6, 92.2, 47.9, 20.7; MS (ESI+) *m/z*: 443.1 (M+H)+ Ana. Calcd for $C_{27}H_{20}O_4$: C, 79.40; H, 4.94; Found: C, 79.45; H, 4.97.



2-(Biphenylcarbonyl)-3-(furan-2-yl)-6-methyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6e) : m.p:125-130°C; as white solid; Yield: 90% ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 8.05 (d, *J* = 8.3Hz, 2H, ArH), 7.74 (d, *J* = 8.3Hz, 2H, Ar-H), 7.65 (d, J = 7.0Hz, 2H, ArH), 7.51-7.43 (m, 4H, ArH), 6.38 (s, 1H, ArH), 6.31 (d, *J* = 2.8Hz, 1H, ArH), 6.20 (d, *J* = 4.6Hz, 1H, CH), 6.14 (s, 1H, CH), 4.86 (d, *J* = 4.4Hz, 1H, CH), 2.30 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 191.6, 171.6, 167.0, 160.8, 151.3, 147.2, 142.8, 139.5, 131.8, 129.8, 129.1, 128.7, 127.7, 127.4, 111.0, 108.2, 99.6, 95.7, 88.9, 41.9, 20.7; MS (ESI^+) *m/z*: 399.1 (M+H)+ Ana. Calcd for $C_{25}H_{18}O_5$: C, 75.37; H, 4.55; Found: C, 75.39; H, 4.50.

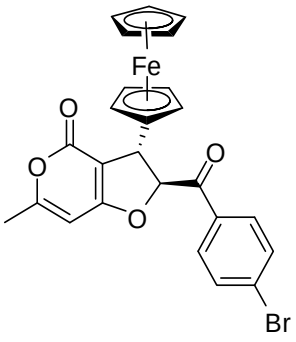


2-(4-Methoxybenzoyl)-6-methyl-3-(naphthalen-1-yl)-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6f): m.p:160-165°C; White solid; yield: 80%; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.96 (d, *J* = 8.0Hz, 1H, Ar-H), 7.86-7.79 (m, 4H, ArH), 7.47-7.41 (m, 3H, ArH), 7.33 (d, *J* = 6.9Hz, 1H, ArH), 6.87 (d, *J* = 8.8Hz, 2H, Ar-H), 6.12 (s, 1H, CH), 5.90 (s, 1H, CH), 5.73 (s, 1H, CH), 3.84 (s, 3H, OCH_3), 2.30 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ =190.8, 170.9, 166.5, 164.5, 161.0, 135.9, 134.2, 131.9, 131.3, 129.0, 128.5, 126.6, 125.9, 125.7, 123.2, 114.1, 102.3, 95.5, 91.8, 60.4, 55.6, 42.6, 20.6, 14.2;



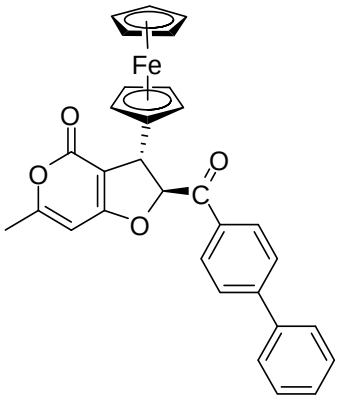
MS (ESI+) *m/z*: 413.1 (M+H)+ Ana. Calcd for $C_{26}H_{20}O_5$: C, 75.72; H, 4.89; Found: C, 75.78; H, 4.85.

2-(4-Bromobenzoyl)-3-ferrocenyl-6-methyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one (6g): m.p: 142-145°C; yellow solid; yield: 89%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.95 (d, *J* = 8.5Hz, 2H, Ar-H), 7.71 (d, *J* = 8.4Hz, 2H, ArH), 6.08 (d, *J* = 3.9Hz, 1H, CH), 6.01 (s, 1H, CH), 4.67 (d, *J* = 3.7Hz, 1H, CH), 4.26 (s, 1H, Fc-H), 4.21-4.12 (m, 8H, Fc-H), 2.27 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 192.3, 169.8, 166.3, 161.0, 132.8, 132.4, 130.7, 129.6, 102.8, 95.4, 91.3, 89.6, 69.6, 69.2, 68.8, 68.5, 67.0, 41.0, 20.6; MS (ESI+) *m/z*: 519.2 (M+H)+ Ana. Calcd for $C_{25}H_{19}FeO_4Br$: C, 57.84; H, 3.69; Found: C, 57.75; H, 3.78.



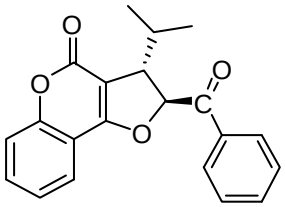
2-(Biphenylcarbonyl)-3-ferrocenyl-6-methyl-2*H*-furo[3,2-c]pyran-4(3*H*)-one(6h):

m.p:175-180°C; yellow solid; yield: 86%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 8.07 (d, *J* = 8.4Hz, 2H, Ar-H), 7.70 (d, *J* = 8.3Hz, 2H, ArH), 7.58 (t, *J* = 6.8Hz, 2H, ArH), 7.43-7.33 (m, 3H, ArH), 6.11 (d, *J* = 3.8Hz, 1H, CH), 5.96 (s, 1H, CH), 4.61 (d, *J* = 3.7Hz, 1H, CH), 4.22-4.05 (m, 9H, Fc-H), 2.18 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 192.7, 170.2, 166.3, 161.3, 147.0, 139.6, 132.7, 129.9, 129.2, 128.7, 127.7, 127.4, 103.0, 95.6, 91.4, 89.1, 68.9, 68.4, 68.1, 67.7, 66.4, 41.3, 20.6; MS (ESI+) *m/z*: 516.1 (M+H)+ Ana. Calcd for $C_{31}H_{24}FeO_4$: C, 72.11; H, 4.68; Found: C, 72.24; H, 4.58.

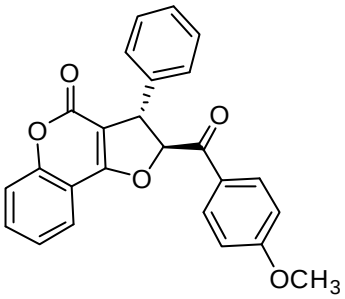


2-Benzoyl-3-isopropyl-2*H*-furo[3,2-c]chromen-4(3*H*)-one (8a):

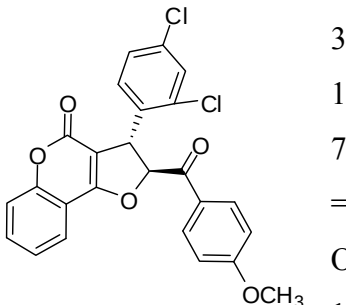
m.p: 98-100°C; white solid; yield: 92%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 8.04 (d, *J* = 7.3Hz, 2H, Ar-H), 7.69-7.51 (m, 4H, ArH), 7.39 (d, *J* = 8.3Hz, 1H, ArH), 7.30 (t, *J* = 7.3Hz, 2H, ArH), 5.94 (d, *J* = 4.6Hz, 1H, CH), 3.90 (t, *J* = 3.9Hz, 1H, CH), 2.54-2.46 (m, 1H, CH), 1.01-0.98 (m, 6H, 2xCH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.7, 166.2, 160.1, 155.0, 134.1, 133.9, 132.6, 129.1, 128.9, 124.0, 122.8, 116.8, 112.0, 103.8, 86.6, 49.0, 29.3, 19.9, 18.1; MS (ESI+) *m/z*: 335.1 (M+H)+ Ana. Calcd for $C_{21}H_{18}O_4$: C, 75.43; H, 5.43; Found: C, 75.40; H, 5.49.



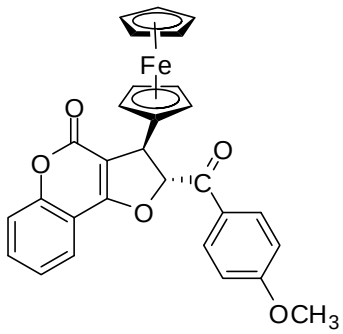
2-(4-Methoxybenzoyl)-3-phenyl-2*H*-furo[3,2-c]chromen-4(3*H*)-one (8b): m.p : 175-180°C; white solid; yield: 87%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.89 (t, *J* = 8.9Hz, 3H, Ar-H), 7.62 (t, *J* = 7.1Hz, 1H, ArH), 7.40-7.32 (m, 7H, ArH), 6.97 (d, *J* = 8.8Hz, 2H, ArH), 6.13 (d, *J* = 4.9Hz, 1H, CH), 4.81(d, *J* = 4.9Hz, 1H, CH), 3.89 (s, 3H, OCH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 190.7, 166.6, 164.6, 159.5, 155.5, 139.8, 133.0, 131.6, 130.2, 129.3, 128.2, 127.7, 126.1, 124.2, 123.3, 117.1, 114.4, 112.3, 105.5, 92.6, 55.7, 49.6; MS (ESI+) *m/z*: 399.1 (M+H)+ Ana. Calcd for $C_{25}H_{18}O_5$: C, 75.37; H, 4.55; Found: C, 75.45; H, 4.39.



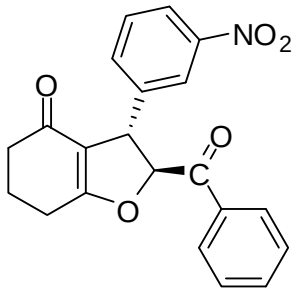
3-(2,4-Dichlorophenyl)-2-(4-methoxybenzoyl)-2*H*-furo[3,2-c]chromen-4(3*H*)-one (8c) : m.p:150-154°C; white solid; yield: 89%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.99 (d, *J* = 8.8Hz, 2H, Ar-H), 7.76 (d, *J* = 6.8Hz, 1H, ArH), 7.63-7.58 (m, 1H, ArH), 7.42 (d, *J* = 8.4Hz, 2H, ArH), 7.34-7.18 (m, 3H, ArH), 6.99 (d, *J* = 8.8Hz, 2H, ArH), 6.07 (d, *J* = 5.1Hz, 1H, CH), 5.45 (d, *J* = 5.0Hz, 1H, CH), 3.90 (s, 3H, OCH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 189.6, 166.8, 164.6, 159.1, 155.4, 135.7, 134.4, 134.3, 133.1, 131.6, 130.4, 130.0, 127.9, 126.5, 124.2, 123.1, 117.1, 114.3, 112.0, 103.9, 90.6, 55.6, 45.3; MS (ESI+) *m/z*: 467.1 (M+H)+ Ana. Calcd for $C_{25}H_{16}Cl_2O_5$: C, 64.26; H, 3.45; Found: C, 64.32; H, 3.22.



3-Ferrocenyl,2-(4-methoxybenzoyl)-2*H*-furo[3,2-c]chromen-4(3*H*)-one (8d): m.p :150-155°C; yellow solid; yield: 88%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 8.16 (d, *J* = 8.6Hz, 2H, Ar-H), 7.71 (d, *J* = 7.6Hz, 1H, ArH), 7.58 (t, *J* = 7.5Hz, 1H, ArH), 7.38 (d, *J* = 8.3Hz, 1H, ArH), 7.29 (d, *J* = 7.9Hz, 1H, ArH), 7.06 (d, *J* = 8.6Hz, 2H, ArH), 6.31 (d, *J* = 3.9Hz, 1H, CH), 4.89 (d, *J* =3.9Hz, 1H, CH), 4.36 (s, 1H, Fc-H), 4.23-4.11 (m, 8H, Fc-H), 3.92 (s, 3H, OCH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 191.2, 165.1, 164.6,159.8, 155.2, 132.7, 131.8, 127.1, 124.1, 123.0, 117.0, 114.4, 112.4, 106.0, 91.5, 88.7, 68.8, 68.4, 68.3, 66.2, 55.8, 42.1; MS (ESI+) *m/z*: 507.1 (M+H)+ Ana. Calcd for $C_{29}H_{22}FeO_5$: C, 68.79; H, 4.38; Found: C, 68.85; H, 4.25.

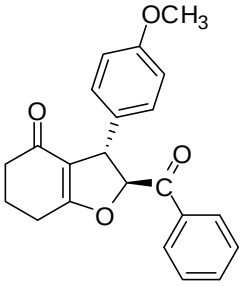


2-Benzoyl-3-(3-nitrophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10a):



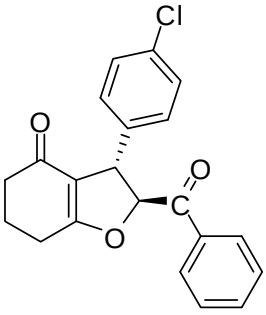
m.p:145-150°C; 88% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 8.13 (t, *J* = 7.9Hz, 2H, ArH), 7.86 (d, *J* = 7.4Hz, 2H, Ar-H), 7.66-7.45 (m, 5H, ArH), 5.85 (d, *J* = 5.1Hz, 1H, CH), 4.68 (d, *J* = 4.4Hz, 1H, CH), 2.72 (s, 2H, CH_2), 2.34-2.32 (m, 2H, CH_2), 2.16-2.12 (m, 2H, CH_2); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 194.2, 192.2, 148.6, 143.1, 134.4, 134.0, 133.2, 129.9, 129.0, 122.6, 122.1, 115.5, 90.9, 47.8, 36.6, 23.8, 21.6; MS (ESI+) *m/z*: 364.1 (M+H)+ Ana. Calcd for $C_{21}H_{17}NO_5$: C, 69.41; H, 4.72; N, 3.85; Found: C, 69.26; H, 4.65; N, 3.79.

2-Benzoyl-3-(4-methoxyphenyl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10b) :

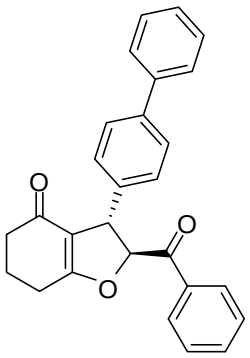


m.p:115-120°C; 85% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.83 (d, *J* = 7.3Hz, 2H, ArH), 7.62 (t, *J* = 7.1Hz, 1H, Ar-H), 7.47 (t, *J* = 7.7Hz, 2H, ArH), 7.16 (d, *J* = 8.5Hz, 2H, ArH), 6.89 (d, *J* = 8.6Hz, 2H, ArH), 5.83 (d, *J* = 4.6Hz, 1H, CH), 4.36 (d, *J* = 3.8Hz, 1H, CH), 3.79 (s, 3H, OCH_3), 2.70 (s, 2H, CH_2), 2.34-2.29 (m, 2H, CH_2), 2.12-2.08 (m, 2H, CH_2); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 194.5, 193.1, 177.3, 159.1, 134.2, 133.4, 129.0, 128.5, 116.7, 114.5, 91.8, 55.4, 48.5, 36.9, 29.8, 24.0, 21.8; MS (ESI+) *m/z*: 349.1 (M+H)+ Ana. Calcd for $C_{22}H_{20}O_4$: C, 75.84; H, 5.79; Found:C, 75.85; H, 5.70.

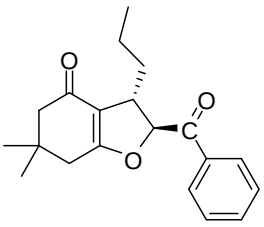
2-Benzoyl-3-(4-chlorophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10c): m.p:160-165°C; 89% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.82 (d, *J* = 7.6Hz, 2H, ArH), 7.63 (t, *J* = 7.0Hz, 1H, Ar-H), 7.47 (t, *J* = 7.2Hz, 2H, ArH), 7.31 (d, *J* = 7.6Hz, 2H, ArH), 7.17 (d, *J* = 7.8Hz, 2H, ArH), 5.81 (d, *J* = 4.6Hz, 1H, CH), 4.43 (d, *J* = 3.6Hz, 1H, CH), 2.69 (s, 2H, CH_2), 2.33-2.29 (m, 2H, CH_2), 2.14-2.12 (m, 2H, CH_2); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 194.06, 192.48, 177.38, 139.59, 134.13, 133.12, 128.98, 128.80, 128.65, 115.92, 91.12, 48.01, 36.55, 23.70, 21.53; MS (ESI+) *m/z*: 353.1(M+H)+ Ana. Calcd for $C_{21}H_{17}ClO_3$: C, 71.49; H, 4.86; Found:C, 71.55; H, 4.81.



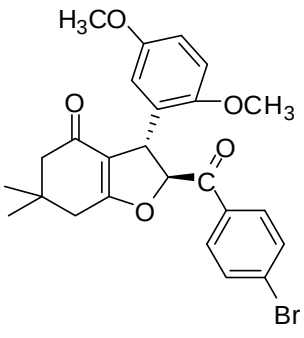
2-Benzoyl-3-(biphenyl-4-yl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10d) : m.p:155-158°C; 82% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.82 (d, *J* = 7.4Hz, 2H, ArH), 7.63 (t, *J* = 7.3Hz, 1H, Ar-H), 7.47-7.32 (m, 7H, ArH), 7.16 (d, *J* = 8.5Hz, 2H, ArH), 6.96 (d, *J* = 8.5Hz, 2H, ArH), 5.83 (d, *J* = 4.6Hz, 1H, CH), 4.37 (d, *J* = 4.0Hz, 1H, CH), 2.70 (s, 2H, CH_2), 2.34-2.30 (m, 2H, CH_2), 2.12-2.08 (m, 2H, CH_2); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 194.4, 193.0, 177.3, 158.3, 137.0, 134.2, 133.6, 133.3, 129.0, 129.0, 128.7, 128.5, 128.1, 127.6, 116.6, 115.4, 91.8, 70.1, 48.4, 36.8, 23.9, 21.8; MS (ESI+) *m/z*: 395.2 (M+H)+ Ana. Calcd for $C_{27}H_{22}O_3$: C, 82.21; H, 5.62; Found:C, 82.25; H, 5.60.



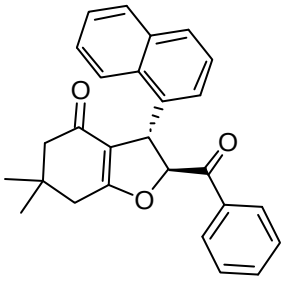
2-Benzoyl-6,6-dimethyl-3-propyl-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10e) : m.p: 94-96°C; White solid; yield: 94%; ^{1}H NMR ($CDCl_3$, 300MHz) δ = 7.93 (d, *J* = 7.3Hz, 2H, ArH), 7.64 (t, *J* = 7.3Hz, 1H, Ar-H), 7.52 (t, *J* = 7.7Hz, 2H, Ar-H), 5.63 (d, *J* = 4.3Hz, 1H, CH), 3.43 (s, 1H, CH), 2.50-2.33 (m, 2H, CH_2), 2.22 (s, 2H, CH_2), 1.88-1.63 (m, 2H, CH), 1.42-1.30 (m, 2H, CH_2), 1.12 (s, 6H, 2xCH_3), 0.95 (t, *J* = 7.2Hz, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.4, 194.1, 176.2, 133.8, 128.8, 128.7, 88.8, 51.2, 42.9, 37.5, 35.2, 34.1, 28.9, 28.3, 19.4, 14.1; MS (ESI+) *m/z*: 313.2(M+H)+ Ana. Calcd for $C_{20}H_{24}O_3$: C, 76.89; H, 7.74; Found: C, 76.67; H, 7.65.



2-(4-Bromobenzoyl)-3-(2,5-dimethoxyphenyl)-6,6-dimethyl-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10f): m.p:147-150°C; 87% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.73 (d, *J* = 8.5Hz, 2H, ArH), 7.59 (d, *J* = 8.4Hz, 2H, Ar-H), 6.76 (s, 2H, ArH), 6.66 (s, 1H, ArH), 5.72 (d, *J* = 5.1Hz, 1H, CH), 4.81 (d, *J* = 4.4Hz, 1H, CH), 3.72 (s, 3H, OCH_3), 3.55 (s, 3H, OCH_3), 2.57-2.43 (m, 2H, CH_2), 2.25 (m, 2H, CH_2), 1.19 (s, 3H, CH_3), 1.15 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.7, 192.4, 176.8, 153.8, 150.9, 132.8, 132.0, 129.2, 115.0, 113.5, 112.7, 111.9, 90.2, 55.7, 51.3, 43.1, 37.8, 34.3, 29.2, 28.4; MS (ESI+) *m/z*: 485.1 (M+H)+ Ana. Calcd for $C_{25}H_{25}BrO_5$: C, 61.86; H, 5.19; Found: C, 61.88; H, 5.11.

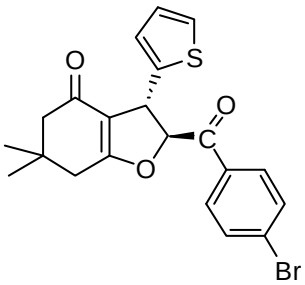


2-Benzoyl-6,6-dimethyl-3-(naphthalen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one

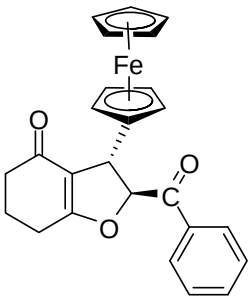


(10g): m.p:175-179°C; 84% as white solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.94-7.77 (m, 5H, ArH), 7.59-7.32 (m, 7H, Ar-H), 5.86 (s, 1H, CH), 5.45 (s, 1H, CH), 2.56 (s, 2H, CH_2), 2.27 (m, 2H, CH_2), 1.22 (s, 3H, CH_3), 1.17 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.7, 193.0, 176.0, 134.2, 133.8, 131.3, 129.4, 128.9, 128.3, 126.4, 125.9, 125.7, 123.4, 115.2, 91.5, 51.4, 37.9, 34.4, 29.2, 28.7; MS (ESI+) *m/z*: 397.1; (M+H)+ Ana. Calcd for $C_{27}H_{24}O_3$: C, 81.79; H, 6.10; Found: C, 81.81; H, 6.14.

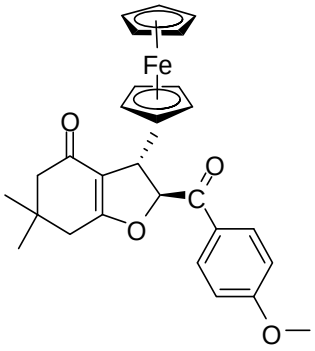
2-(4-Bromobenzoyl)-6,6-dimethyl-3-(thiophen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10h): m.p:145-150°C; White solid, Yield, 86% ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 7.79 (d, *J* = 8.5Hz, 2H, ArH), 7.65 (d, *J* = 8.4Hz, 2H, Ar-H), 7.23 (d, *J* = 4.8Hz, 1H, ArH), 6.98 (dd, *J* = 3.4, 4.5Hz, 2H, ArH), 5.84 (d, *J* = 4.2Hz, 1H, CH), 4.78 (s, 1H, CH), 2.61-2.46 (m, 2H, CH_2), 2.24 (m, 2H, CH_2), 1.19 (s, 3H, CH_3), 1.15 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.4,191.6, 176.4, 144.7, 132.4, 132.0, 130.5, 129.8, 127.4, 125.3, 124.9, 114.5, 91.4, 51.2, 43.8, 37.7, 34.3, 29.2, 28.2; MS (ESI+) *m/z*: 431.0 (M+H)+ Ana. Calcd for $C_{21}H_{19}BrO_3S$: C, 58.47; H, 4.44; Found: C, 58.41; H, 4.46.



2-Benzoyl-3-ferrocenyl-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one(10i):



m.p:120-125°C; 88% as yellow solid; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 8.08 (d, *J* = 7.4Hz, 2H, ArH), 7.67-7.52 (m, 3H, Ar-H), 6.07 (d, *J* = 3.6Hz, 1H, CH), 4.47 (d, *J* = 3.3Hz, 1H, CH), 4.26 (s, 1H, Fc-H), 4.27-4.10 (m, 8H, Fc-H), 2.58-2.56 (m, 2H, CH_2), 2.43-2.26 (m, 2H, CH_2), 2.10-2.01(m, 2H, CH_2); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 194.5, 193.8, 176.1, 134.4, 134.1, 129.2, 129.0, 117.0, 90.5, 90.1, 68.7, 68.0, 66.6, 41.4, 37.0, 24.0, 21.6; MS (ESI+) *m/z*: 427.1 (M+H)+ Ana. Calcd for $C_{25}H_{22}FeO_3$: C, 70.44; H, 5.20; Found: C, 70.24; H, 5.29.

6,6-Dimethyl-3-ferrocenyl-2-(4-methoxybenzoyl)-2,3,6,7-tetrahydrobenzofuran-4(5*H*)-one (10j): 

m.p:124-128°C; yellow solid; yield: 87%; ^{1}H NMR ($CDCl_3$, 300 MHz) δ = 8.07 (d, *J* = 8.8Hz, 2H, ArH), 7.03 (d, *J*= 8.8Hz, 2H, Ar-H), 6.05 (d, *J* =3.7Hz, 1H, CH), 4.43 (d, *J* = 2.6Hz, 1H, CH), 4.20-4.09 (m, 9H, Fc-H), 3.90 (s, 3H, OCH_3), 2.50-2.36 (m, 2H, CH_2), 2.24 (s, 2H), 1.15 (s, 3H, CH_3), 1.12 (s, 3H, CH_3); ^{13}C NMR ($CDCl_3$, 50MHz) δ = 193.9, 192,1, 175.5, 164.3, 131.5, 127.2, 115.4, 114.3, 91.2, 90.5, 68.7, 67.9, 67.8, 66.7, 55.7, 51.5, 41.4, 37.9, 34.1, 29.1, 28.3; MS (ESI+) *m/z*: 485.0 (M+H)+ Ana. Calcd. for $C_{28}H_{28}FeO_4$: C, 69.43; H, 5.83; Found: C, 69.23; H, 5.78;

^{1}H and ^{13}C spectra of all the compounds:

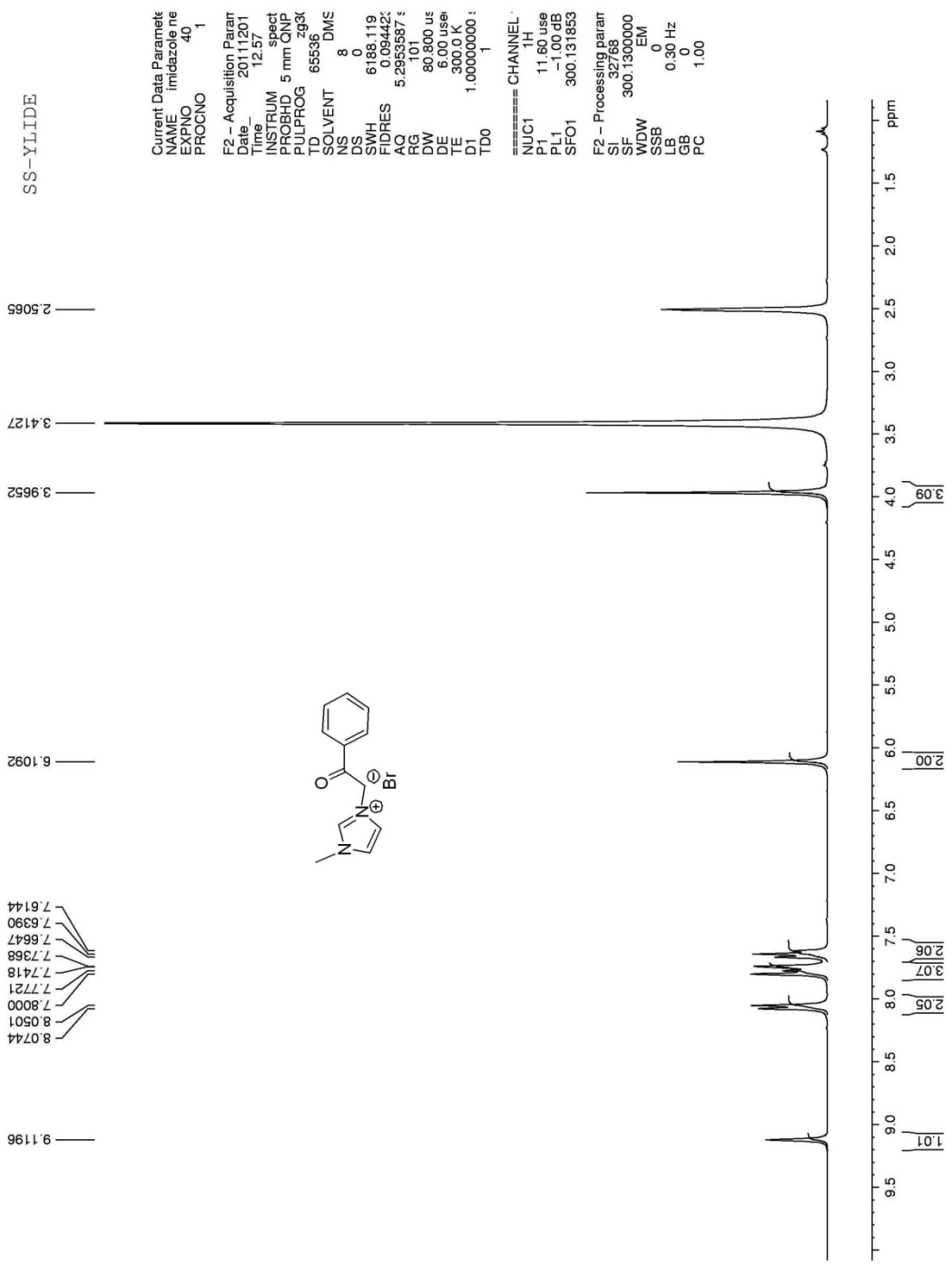


Fig 1: ^{1}H spectra of N-Methyl-3-phenacylimidazolium bromide:

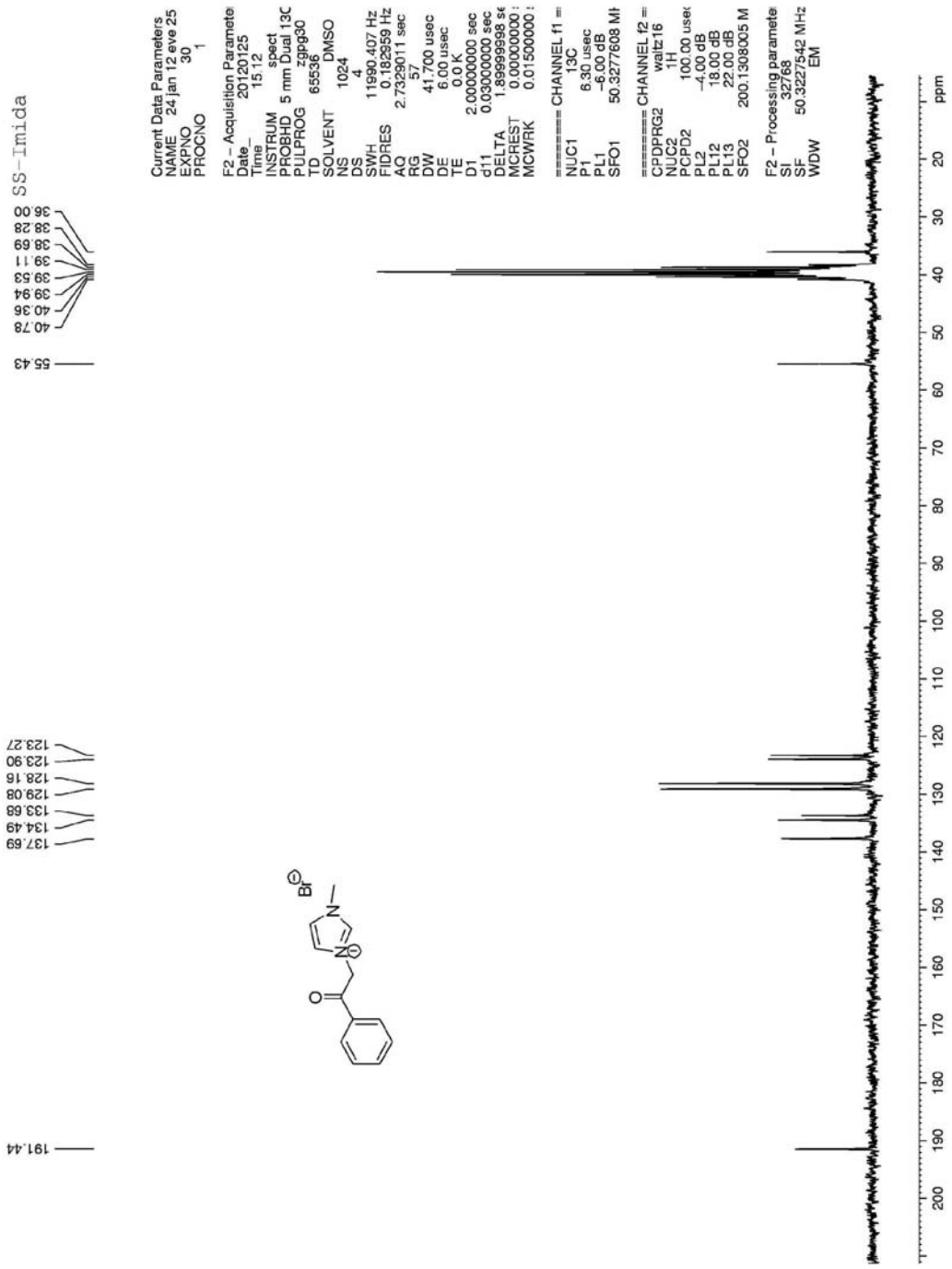


Fig 2: ^{13}C spectra of N-Methyl-3-phenacylimidazolium bromide:

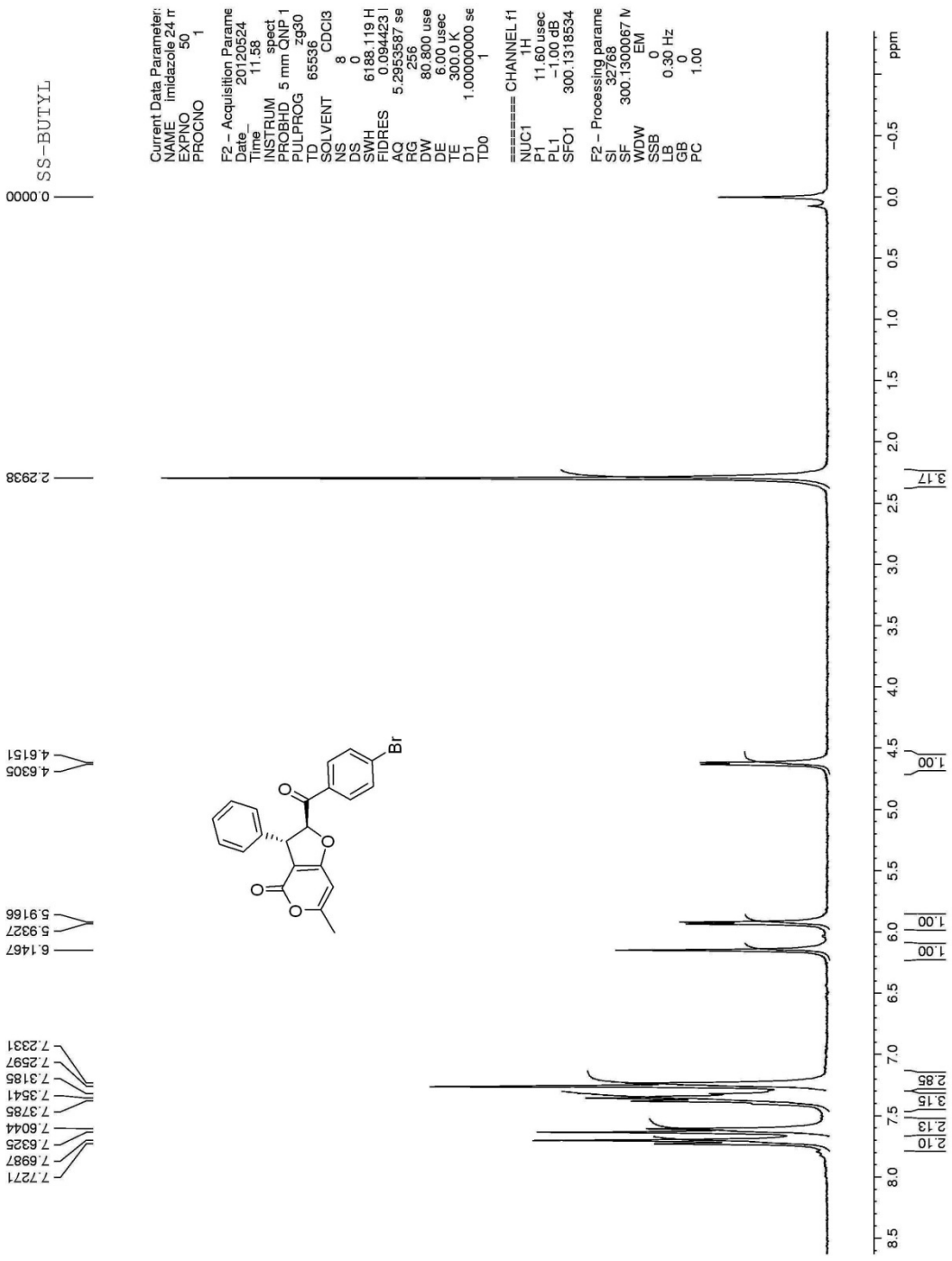


Fig 3: ^{1}H spectra of 2-(4-bromobenzoyl)-6-methyl-3-phenyl-2H-furo[3,2-c]pyran-4(3H)-one:

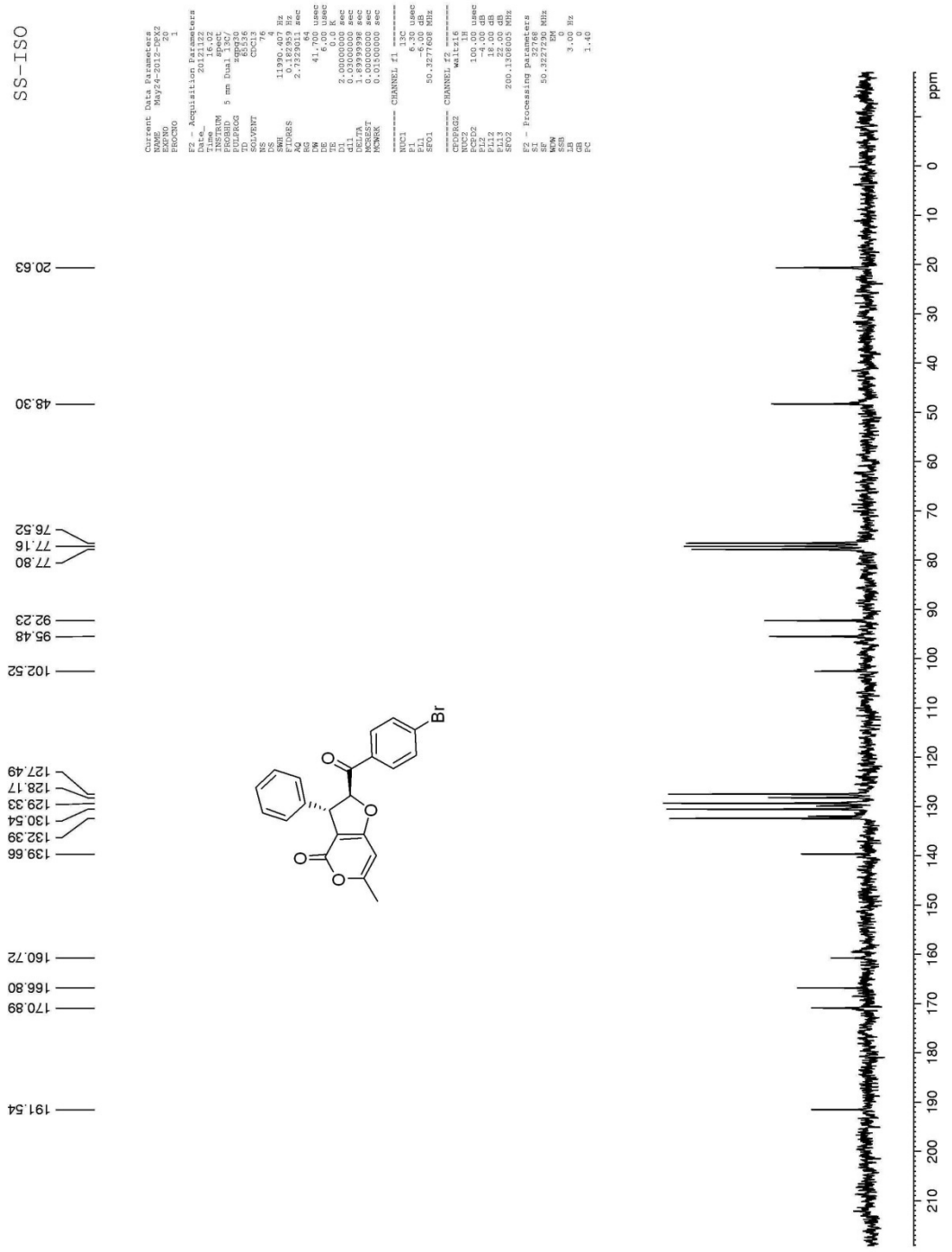


Fig 4: ^{13}C spectra of 2-(4-bromobenzoyl)-6-methyl-3-phenyl-2H-furo[3,2-c]pyran-4(3H)-one :

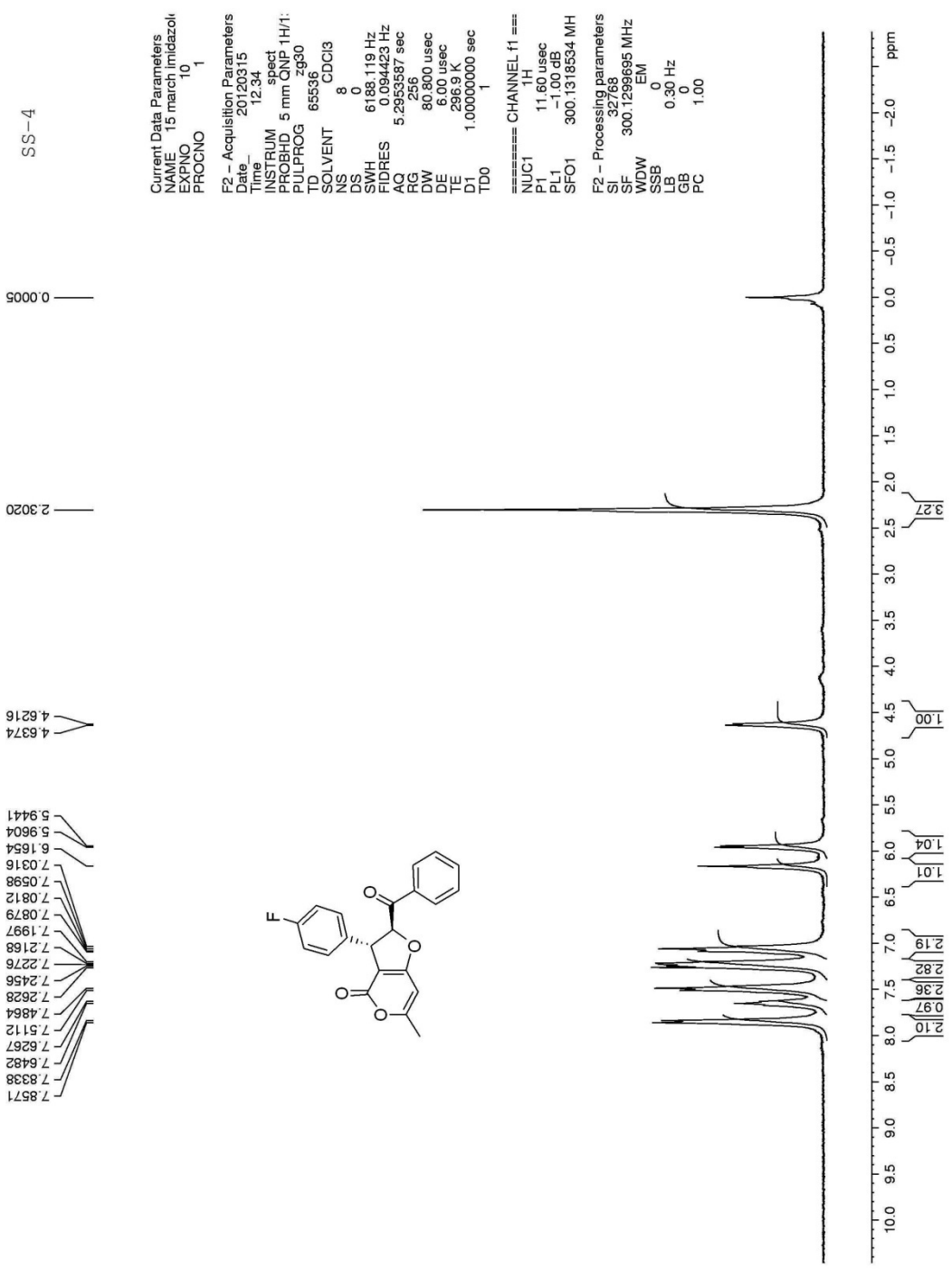


Fig 5: ^{1}H spectra of 2-benzoyl-3-(4-fluorophenyl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one:

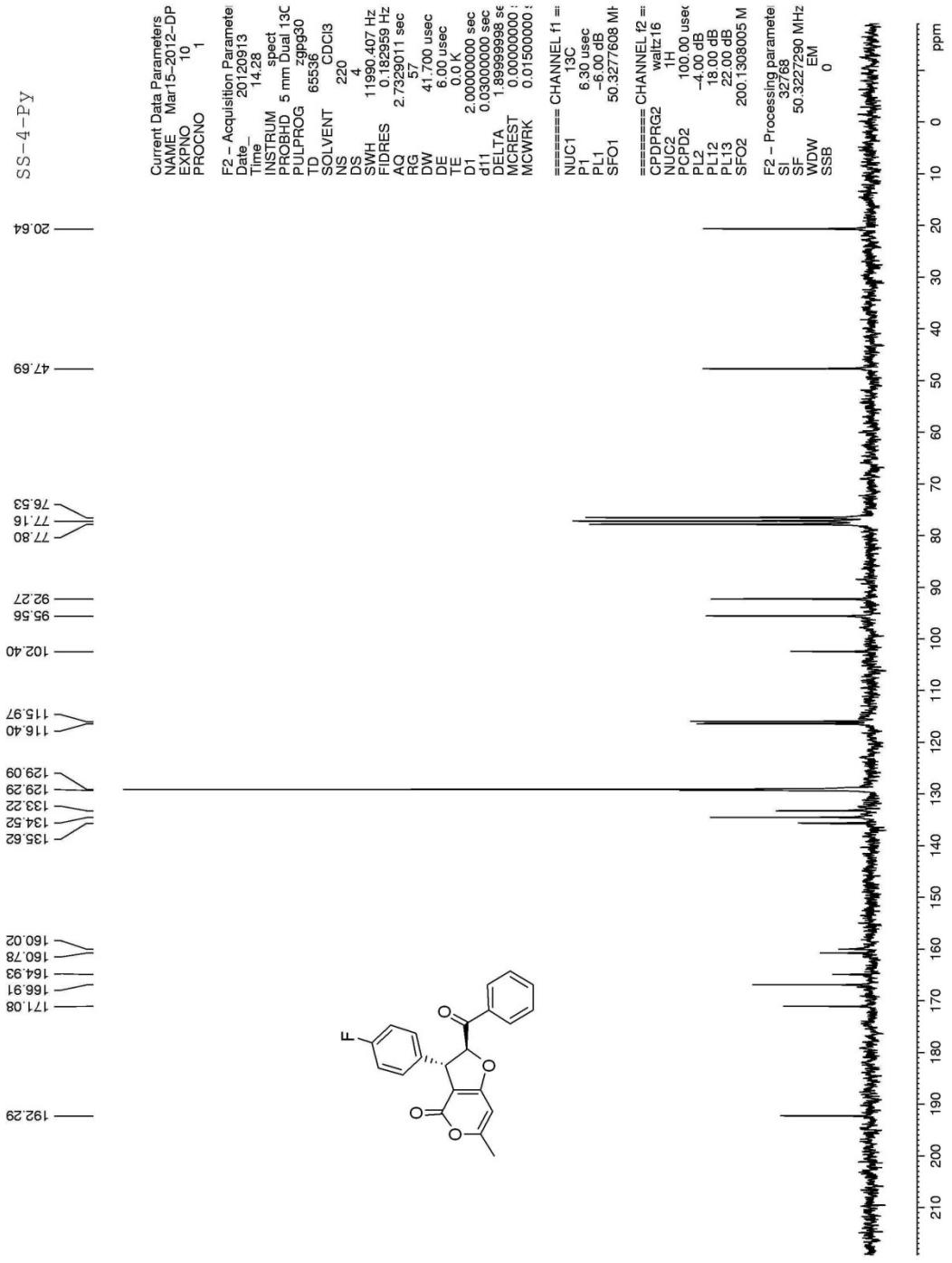


Fig 6: ^{13}C spectra of 2-benzoyl-3-(4-fluorophenyl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one:

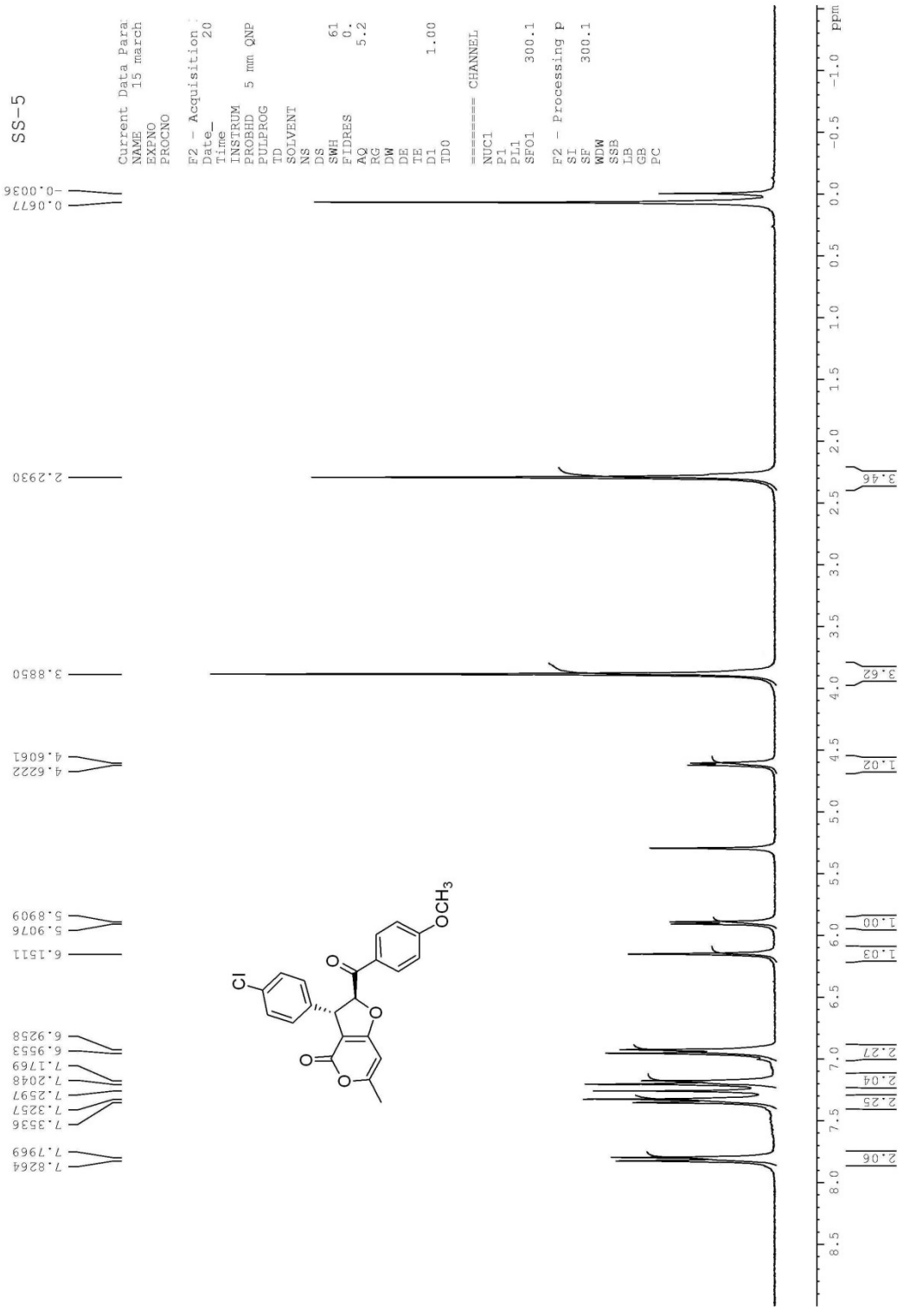


Fig 7: ^{1}H spectra of 3-(4-chlorophenyl)-2-(4-methoxybenzoyl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one

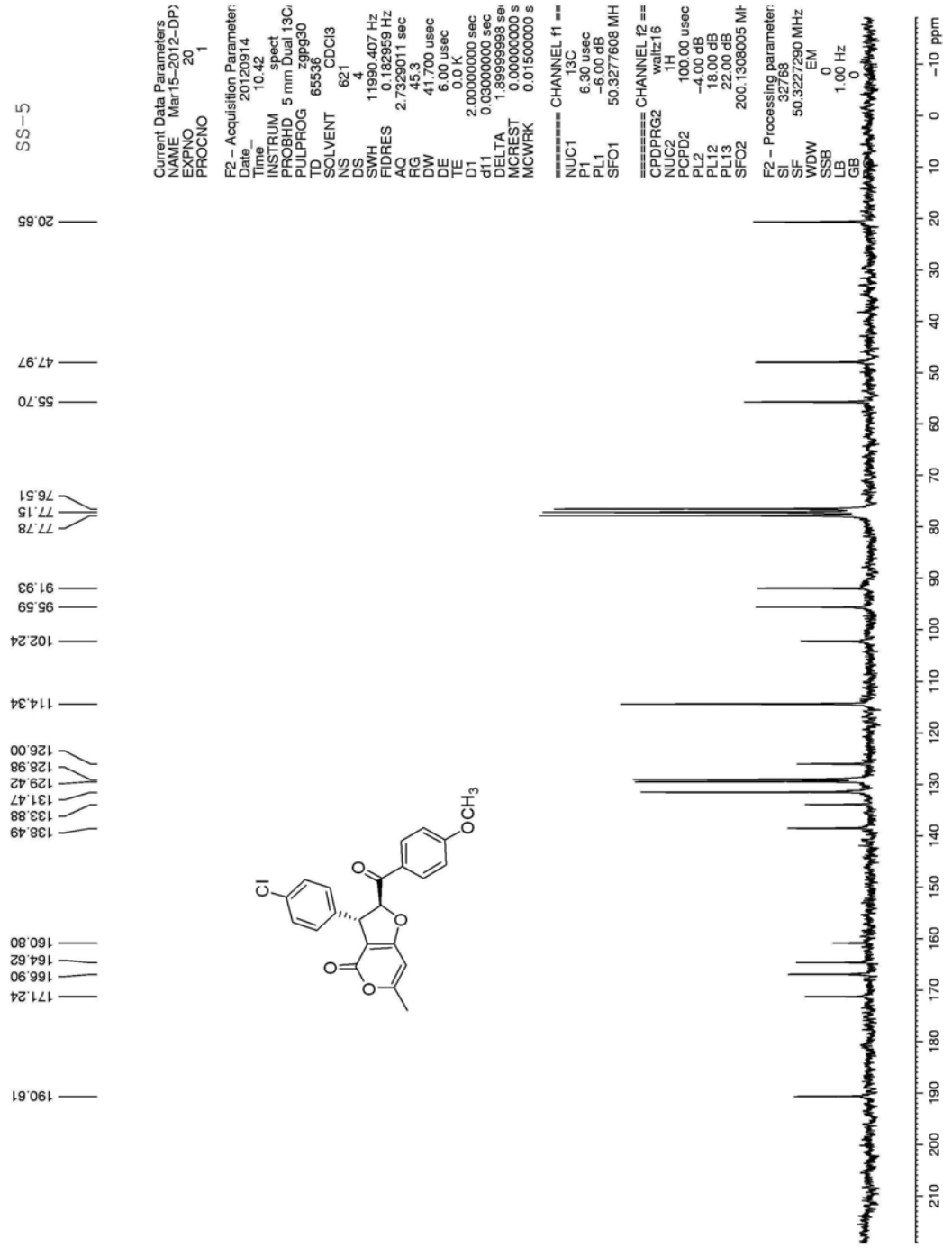


Fig 8: ^{13}C spectra of 3-(4-chlorophenyl)-2-(4-methoxybenzoyl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one

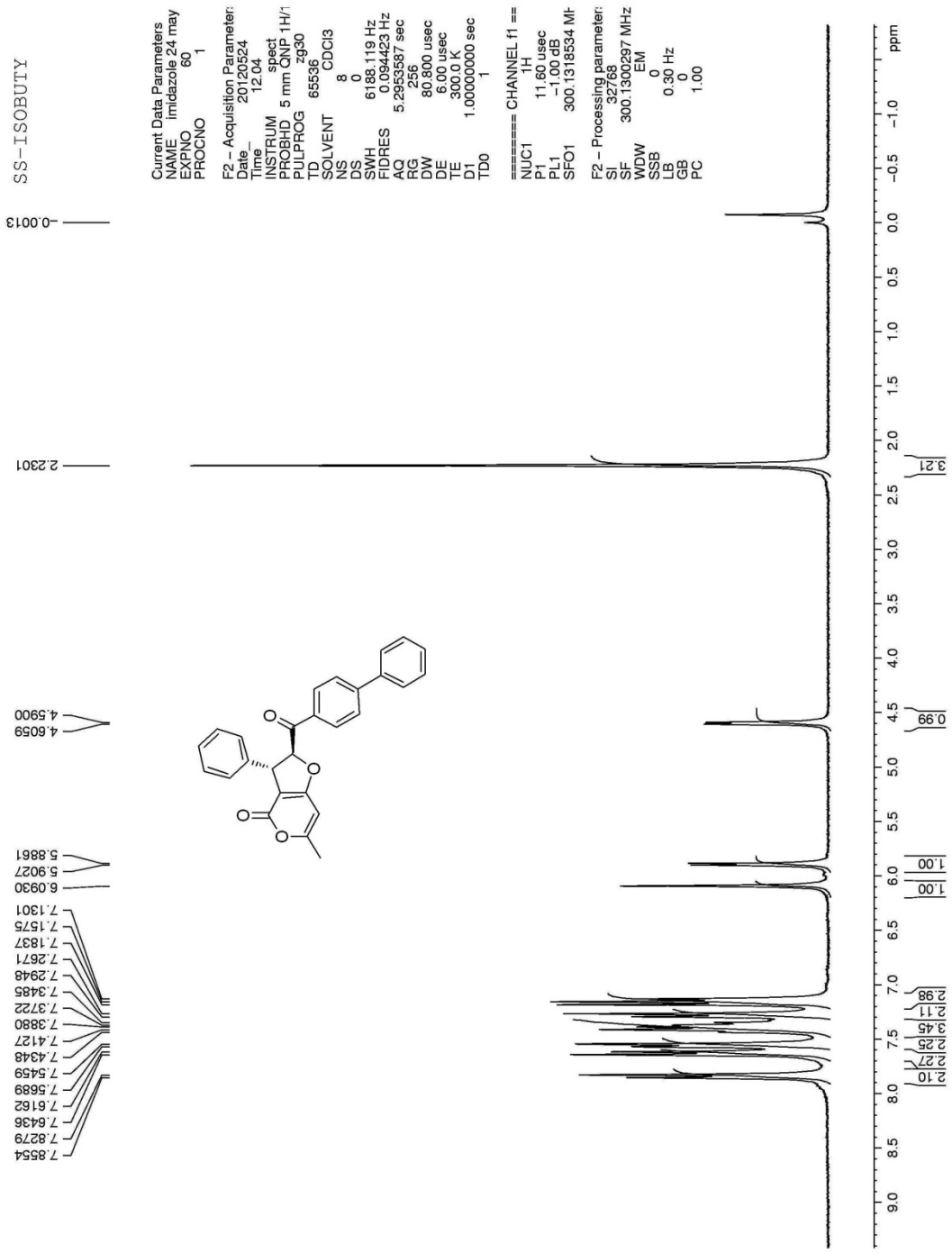


Fig 9:[1]H spectra of 2-(biphenylcarbonyl)-6-methyl-3-phenyl-2H-furo[3,2-c]pyran-4(3H)-one:

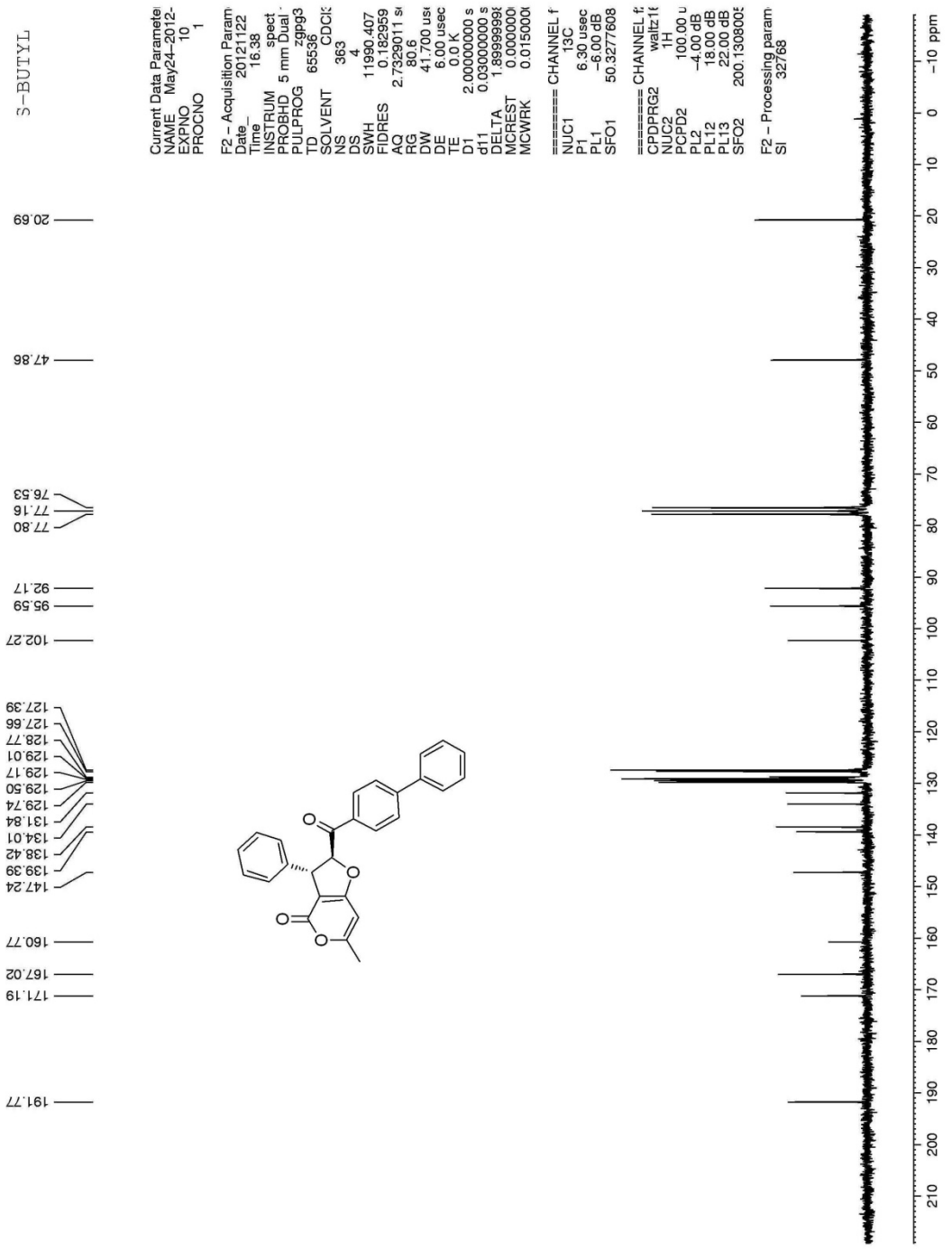


Fig 10:^{13}C spectra of 2-(biphenylcarbonyl)-6-methyl-3-phenyl-2H-furo[3,2-c]pyran-4(3H)-one

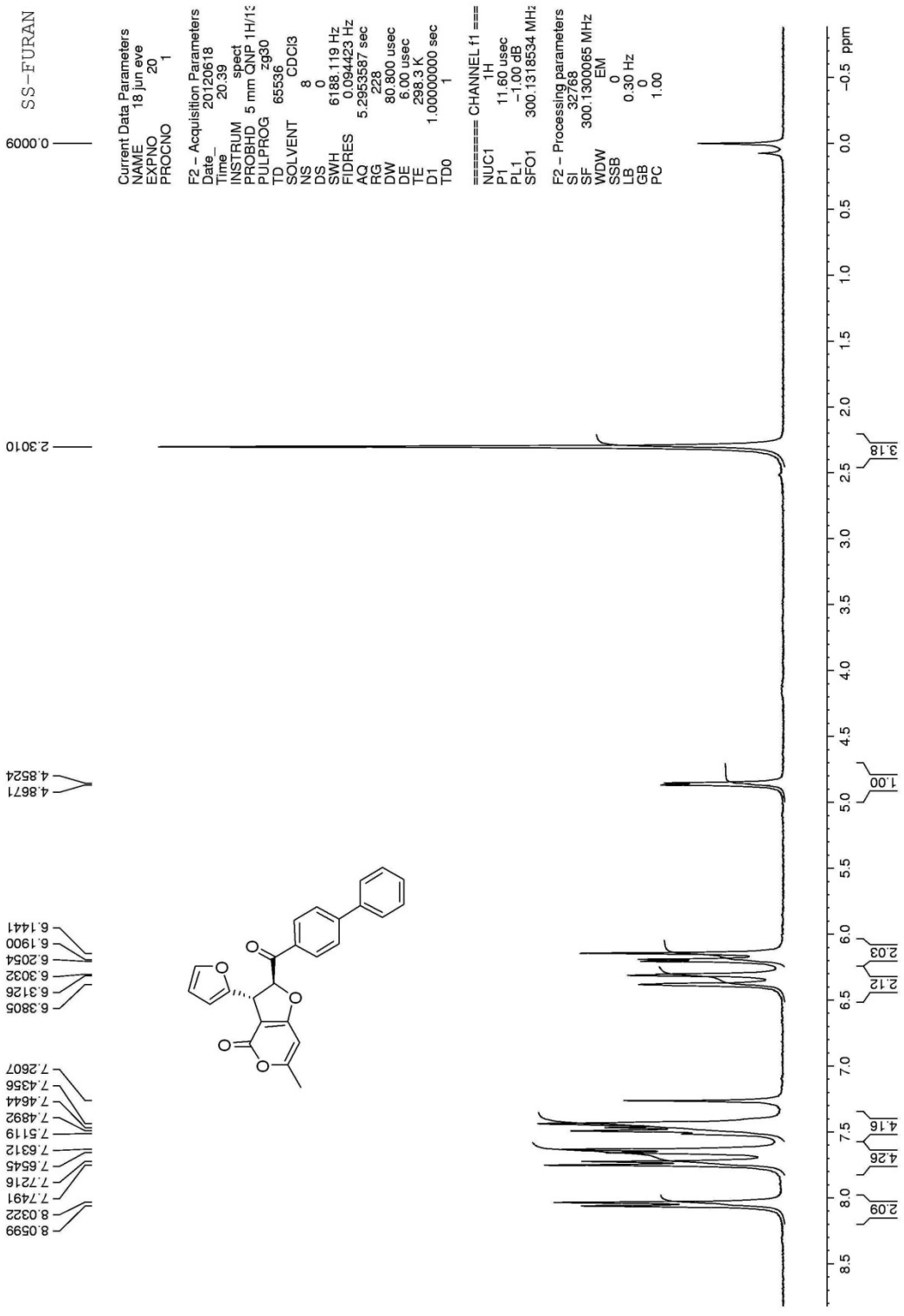


Fig 11:[1]H spectra of 2-(biphenylcarbonyl)-3-(furan-2-yl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one

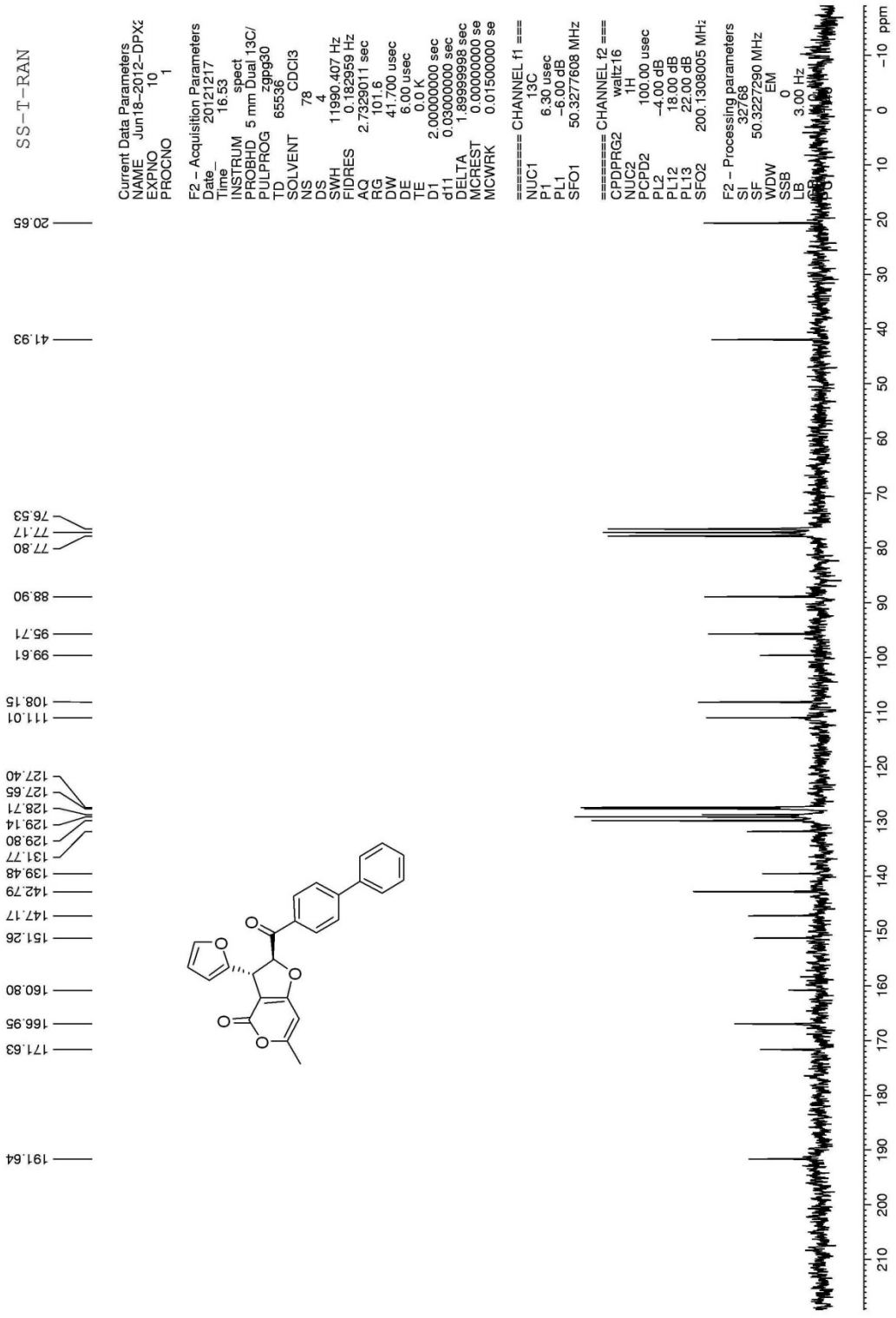


Fig 12: ^{13}C spectra of (2-(biphenylcarbonyl)-3-(furan-2-yl)-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one:

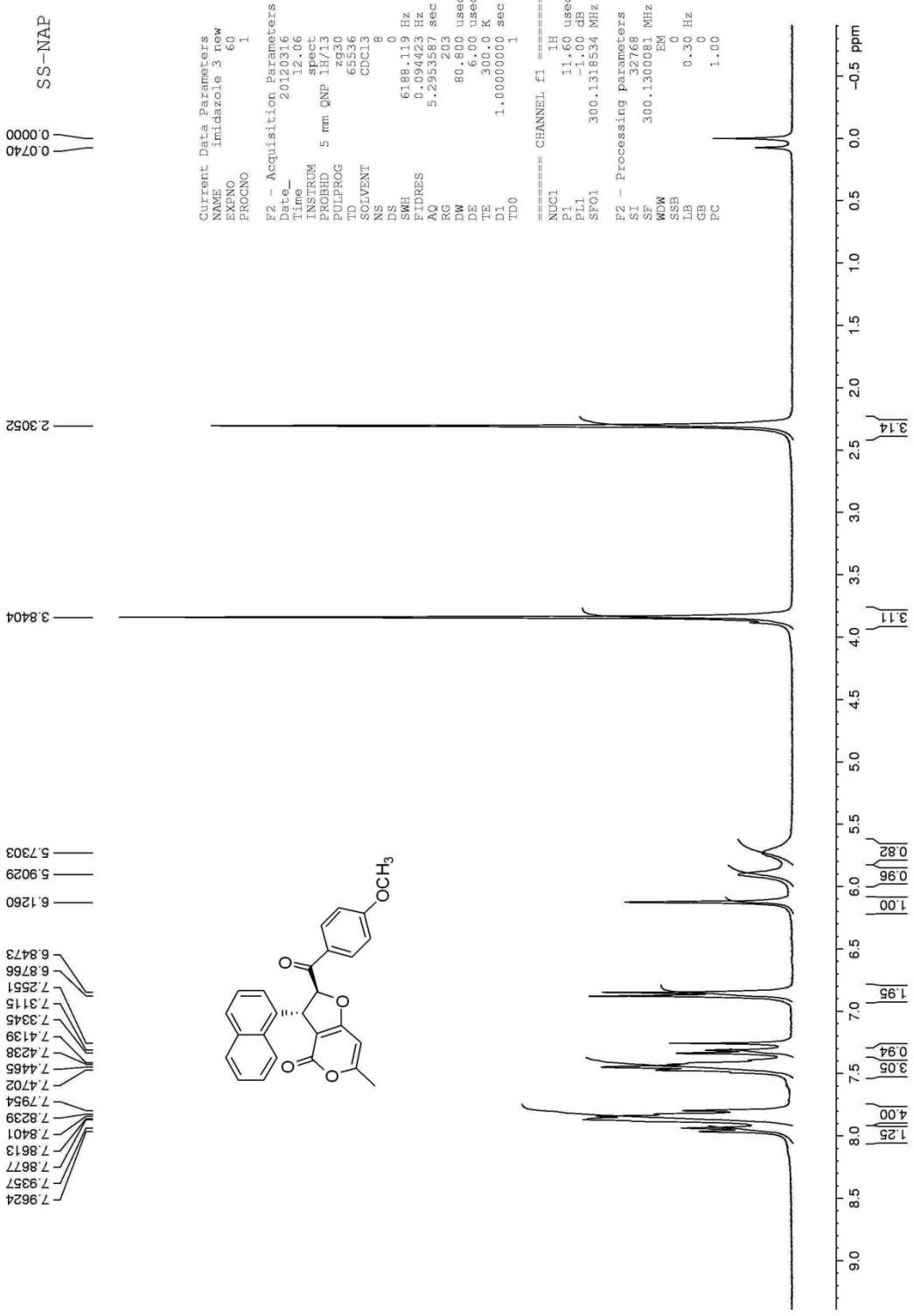


Fig 13:[1]H spectra of 2-(4-methoxybenzoyl)-6-methyl-3-(naphthalen-1-yl)-2H-furo[3,2-c]pyran-4(3H)-one

:

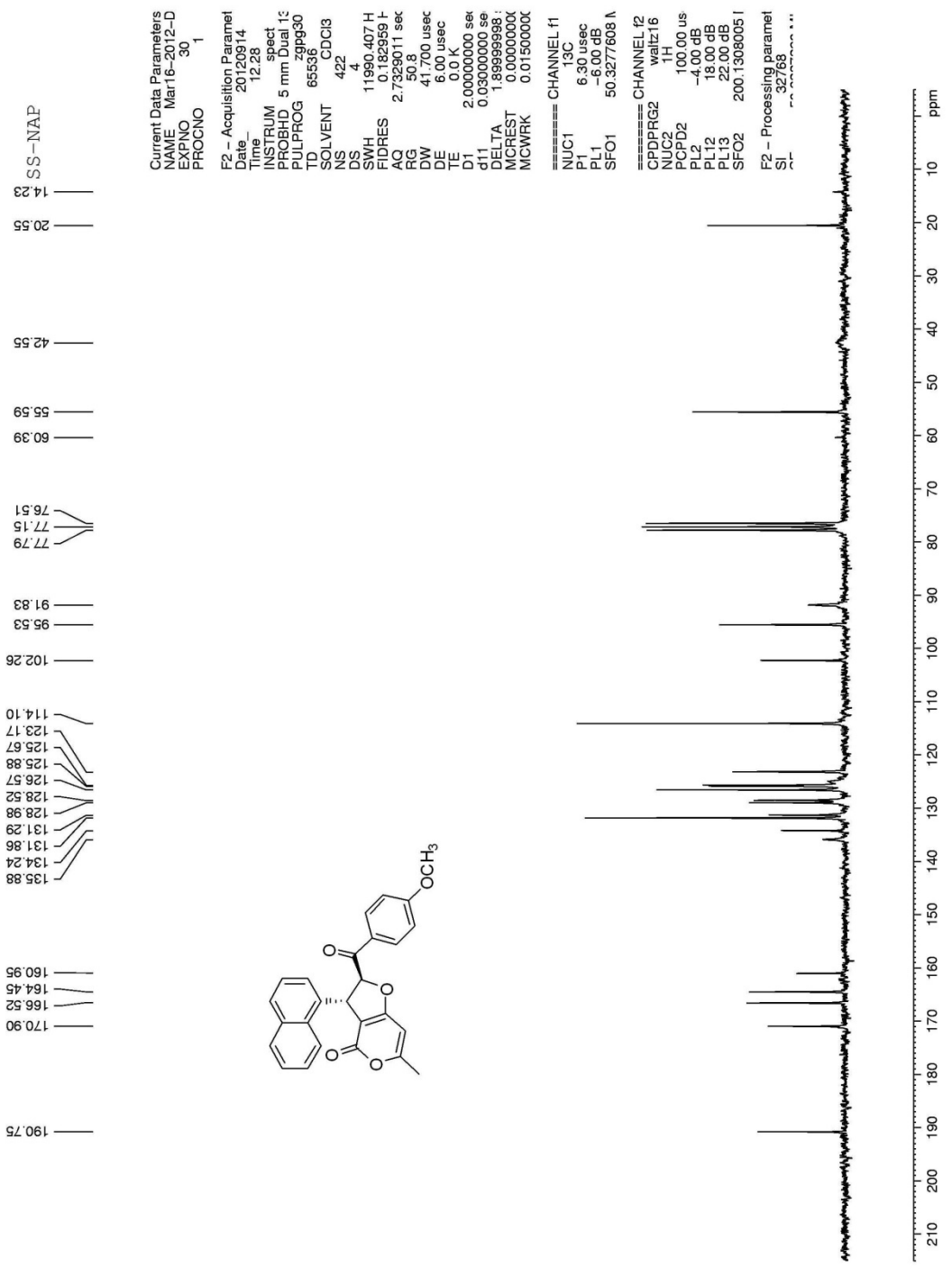


Fig 14: ^{13}C spectra of 2-(4-methoxybenzoyl)-6-methyl-3-(naphthalen-1-yl)-2H-furo[3,2-c]pyran-4(3H)-one:

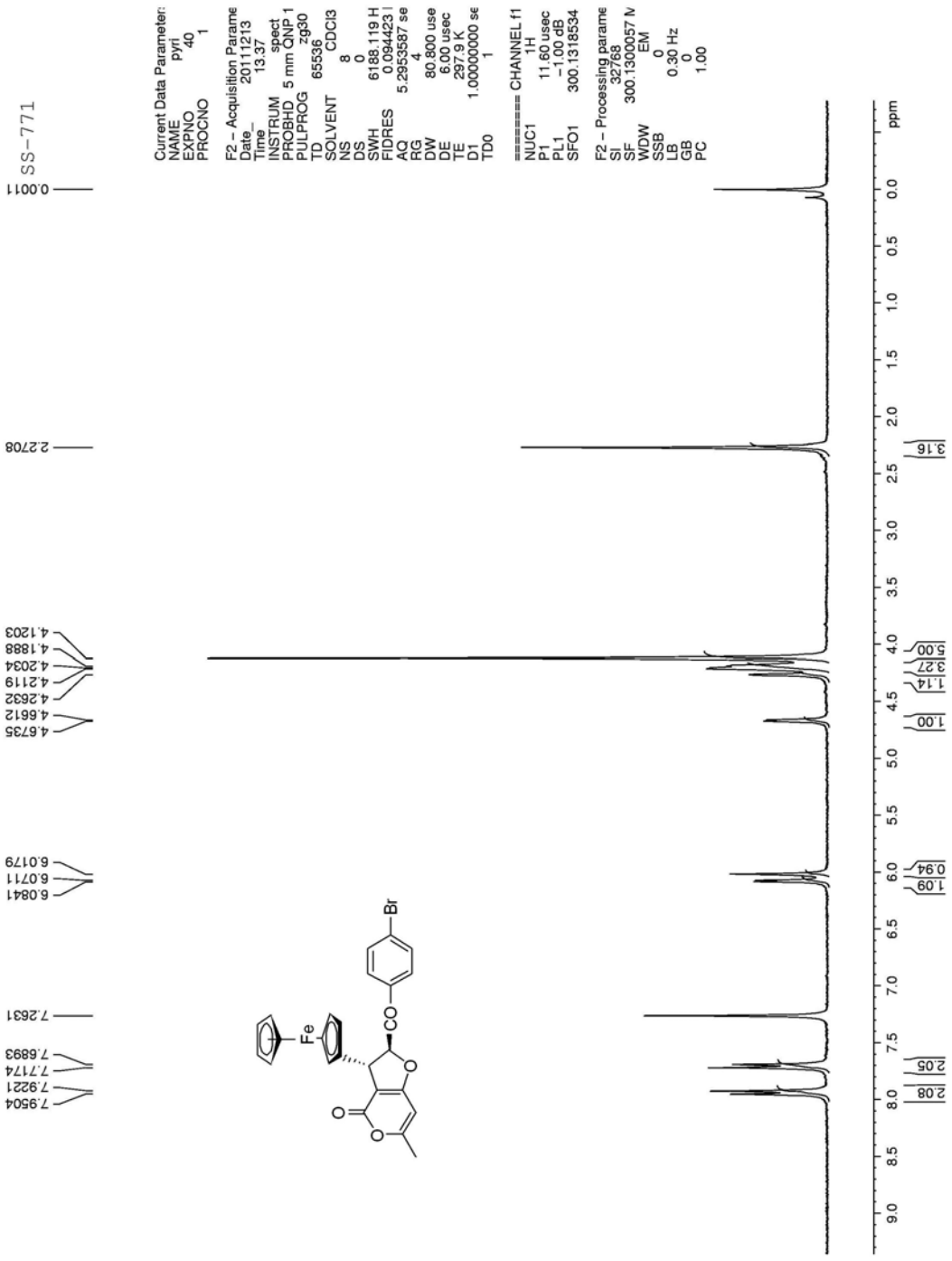


Fig 15: ^{1}H spectra of 2-(4-bromobenzoyl)-3-ferrocenyl-6-methyl- 2H-furo[3,2-c]pyran-4(3H)-one

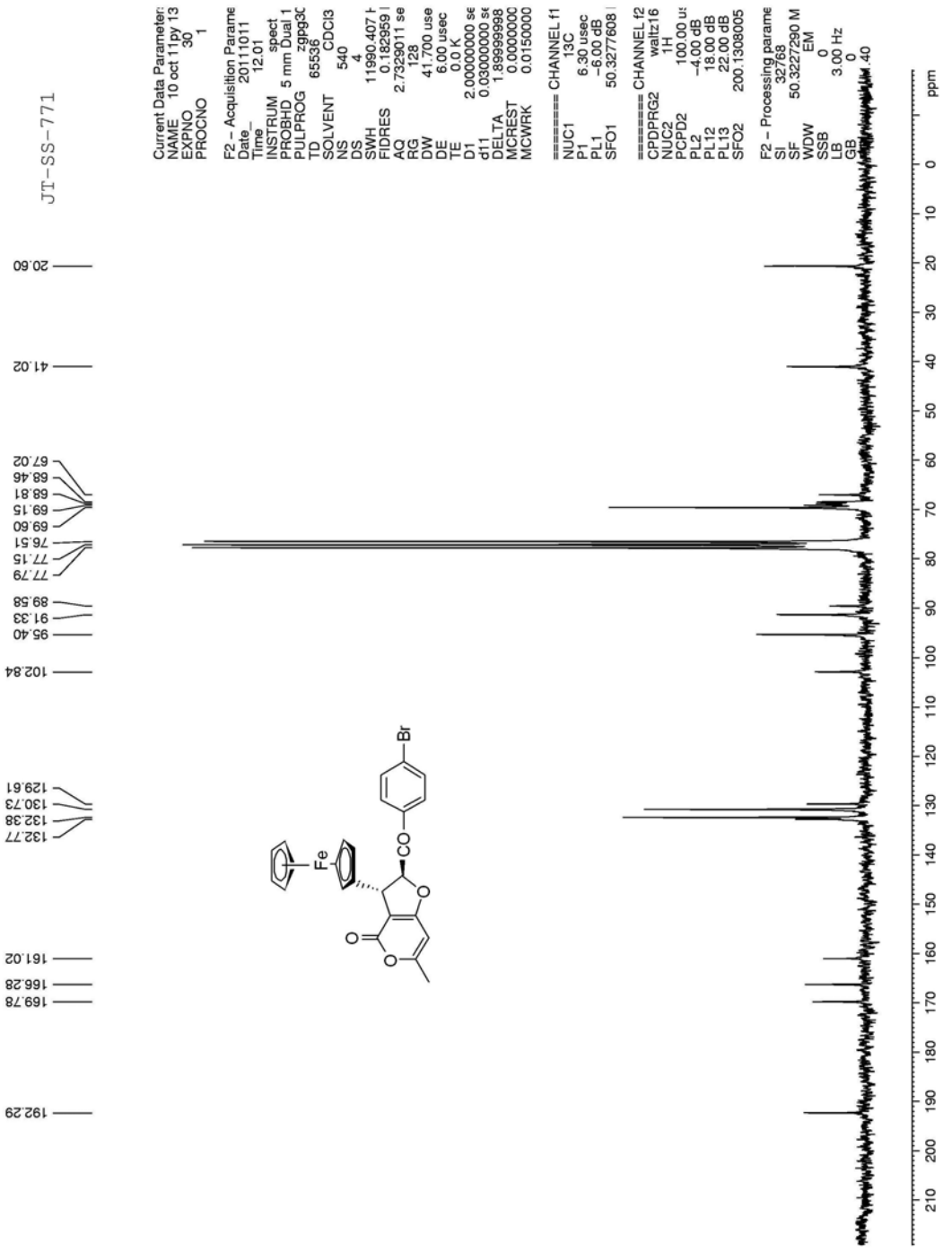


Fig 16: ^{13}C spectra of 2-(4-bromobenzoyl)-3-ferrocenyl-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one

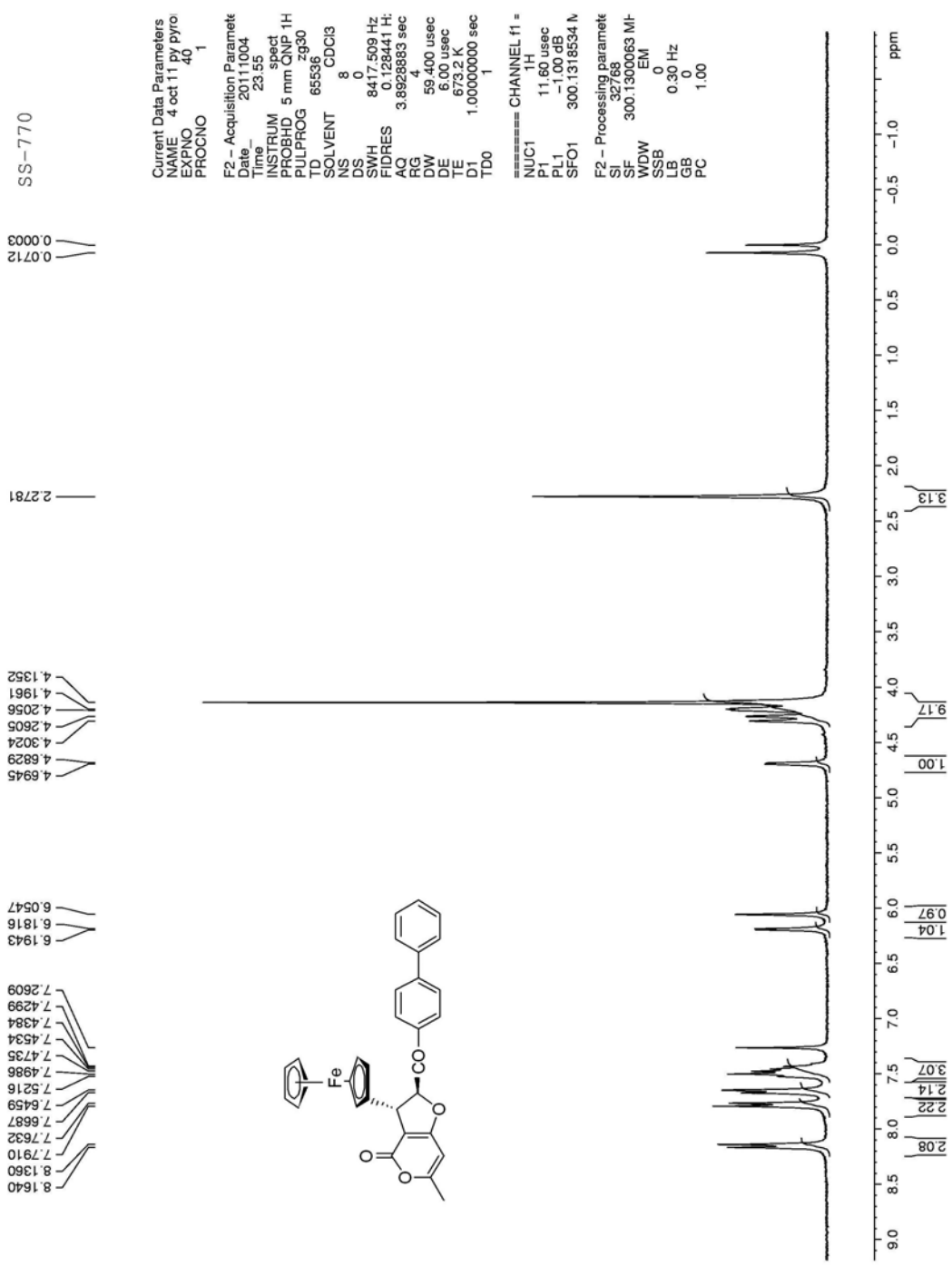


Fig 17: ^{1}H spectra of 2-(biphenylcarbonyl)-3-ferrocenyl-6-methyl-2H-furo[3,2-c]pyran-4(3H)-one

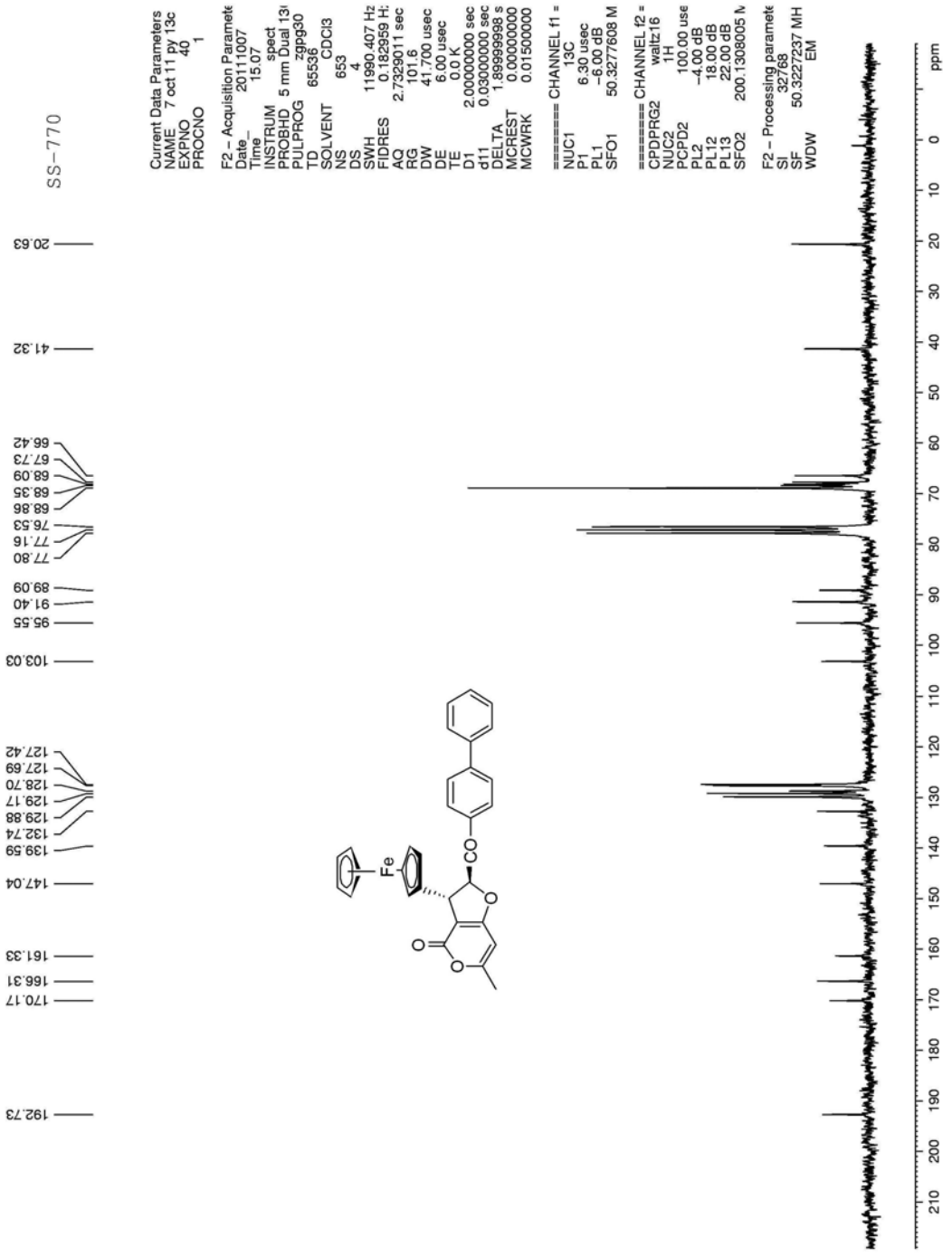


Fig 18: ^{13}C spectra of 2-(biphenylcarbonyl)-3-ferrocenyl 6-methyl- -2H-furo[3,2-c]pyran-4(3H)-one

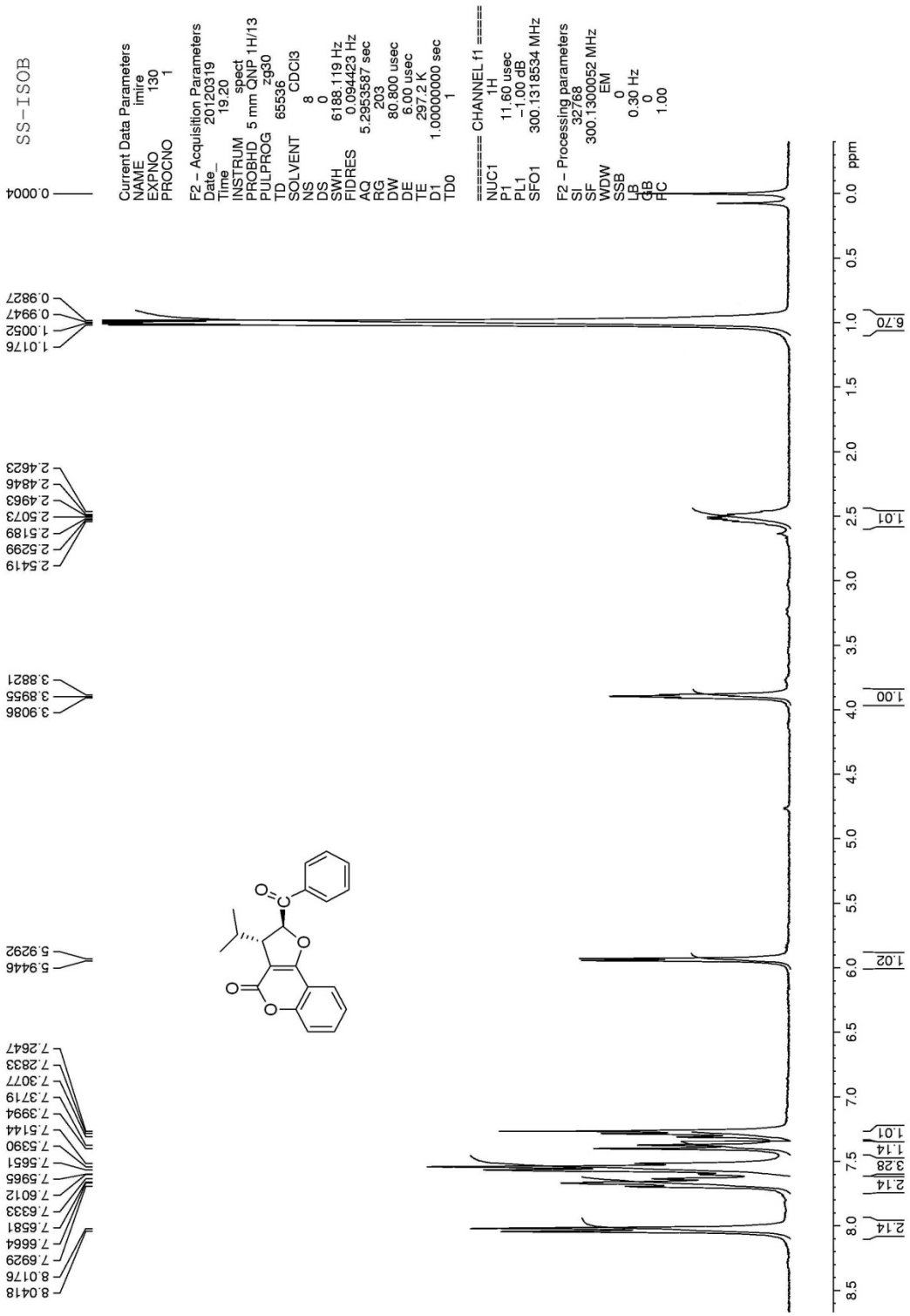


Fig 19: ^{1}H spectra of 2-benzoyl-3-isopropyl-2H-furo[3,2-c]chromen-4(3H)-one:

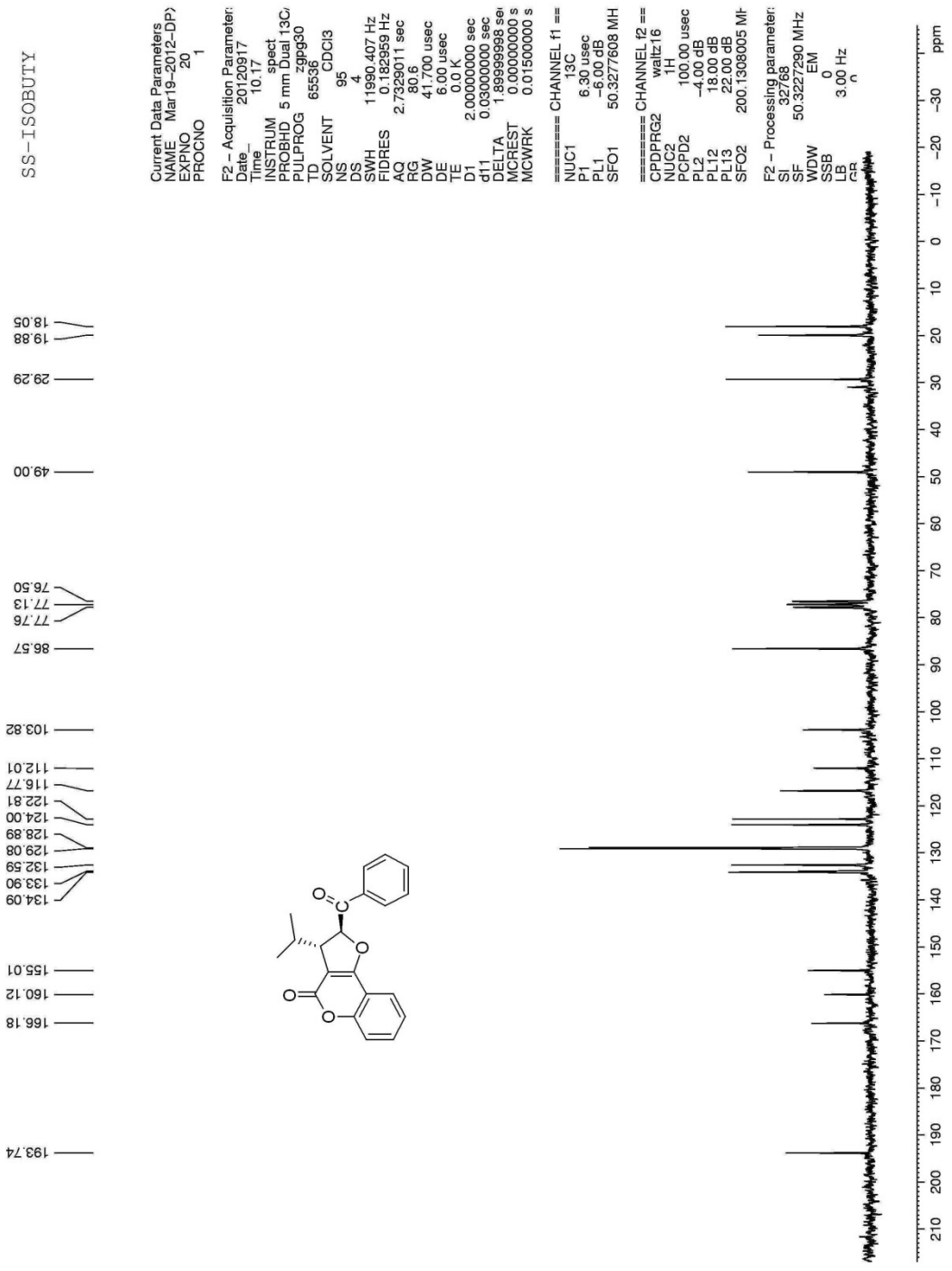


Fig 20: ^{13}C spectra of 2-benzoyl-3-isopropyl-2H-furo[3,2-c]chromen-4(3H)-one:

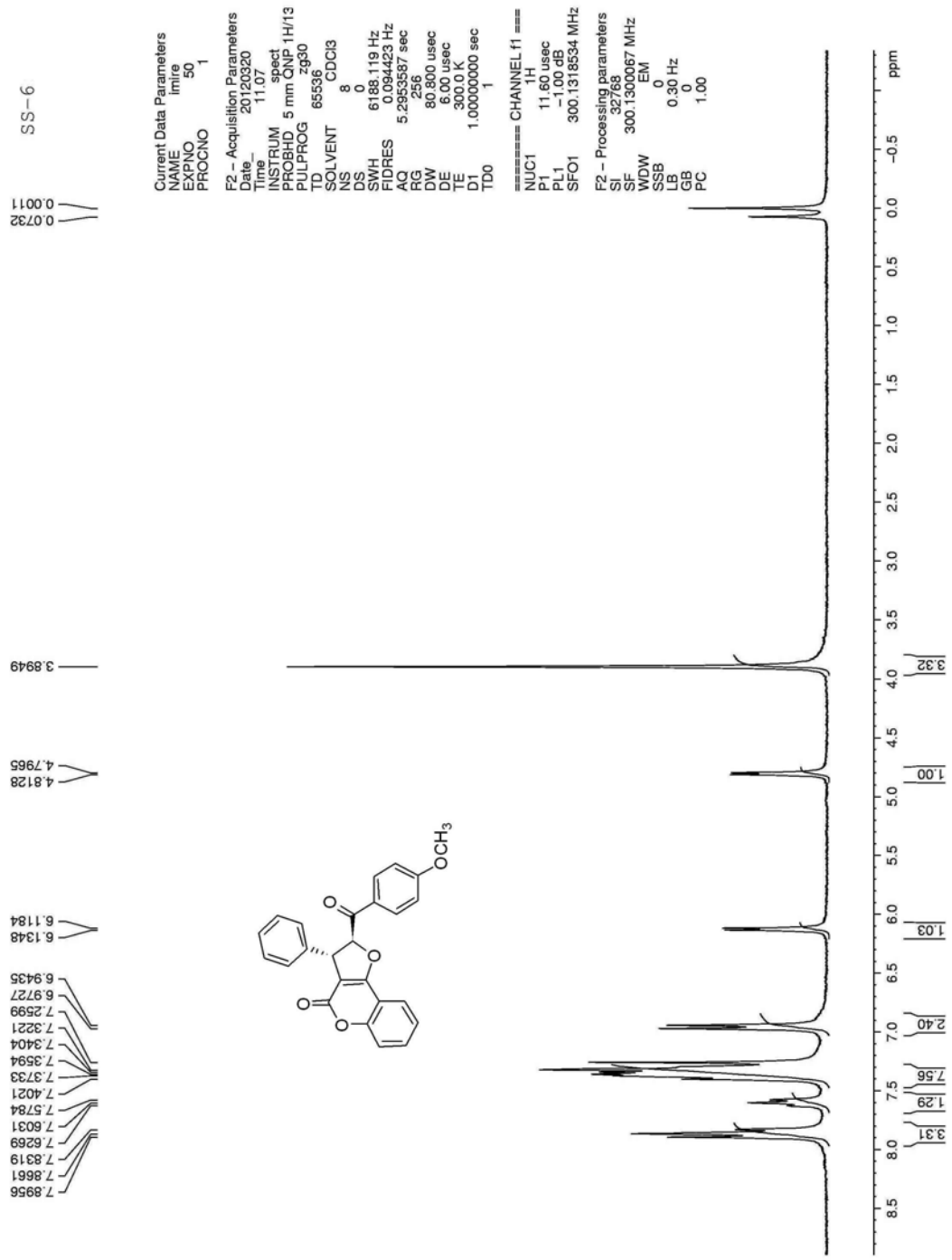


Fig 21: ^{1}H spectra of 2-(4-methoxybenzoyl)-3-phenyl-2H-furo[3,2-c]chromen-4(3H)-one

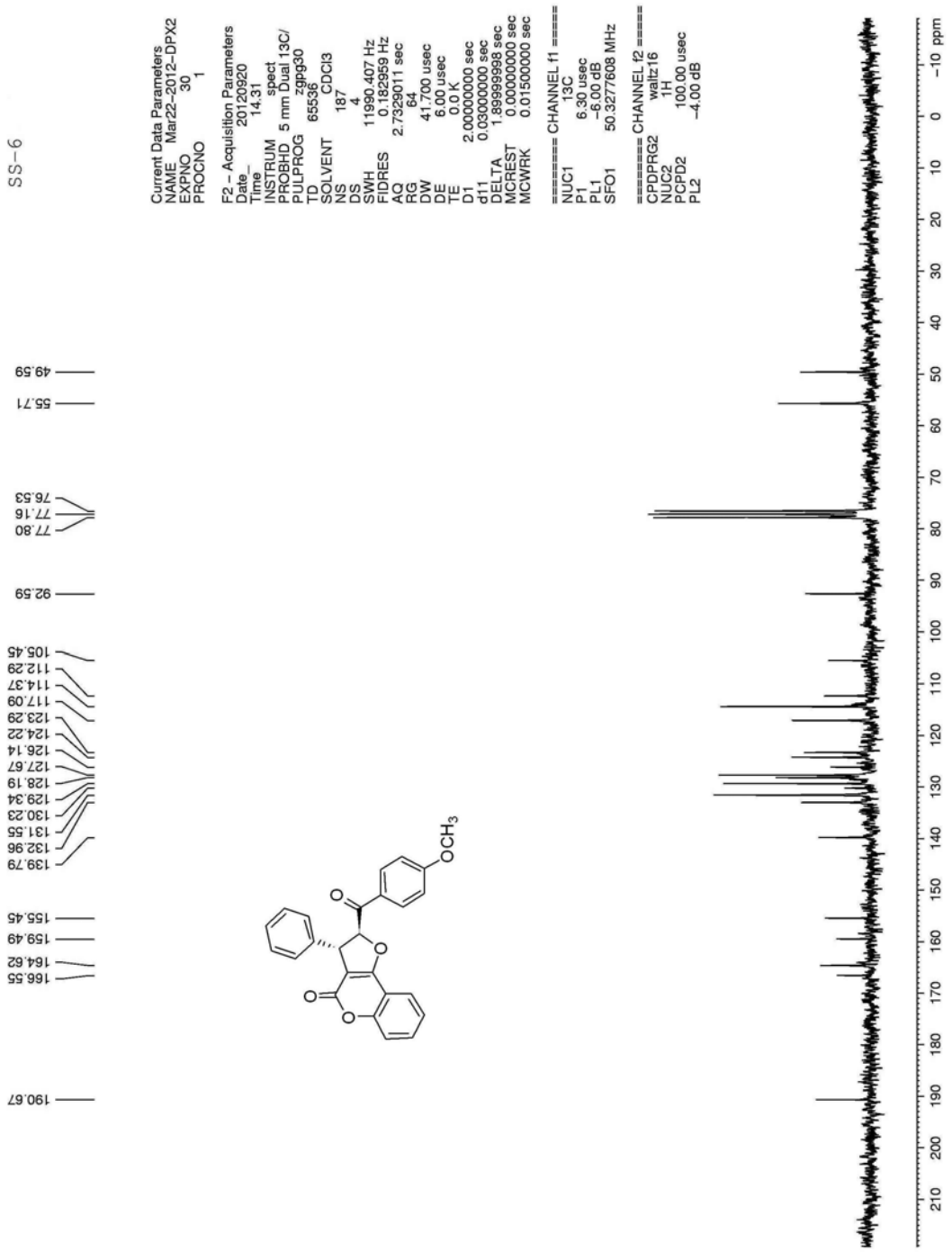


Fig 22: ^{13}C spectra of 2-(4-methoxybenzoyl)-3-phenyl-2H-furo[3,2-c]chromen-4(3H)-one

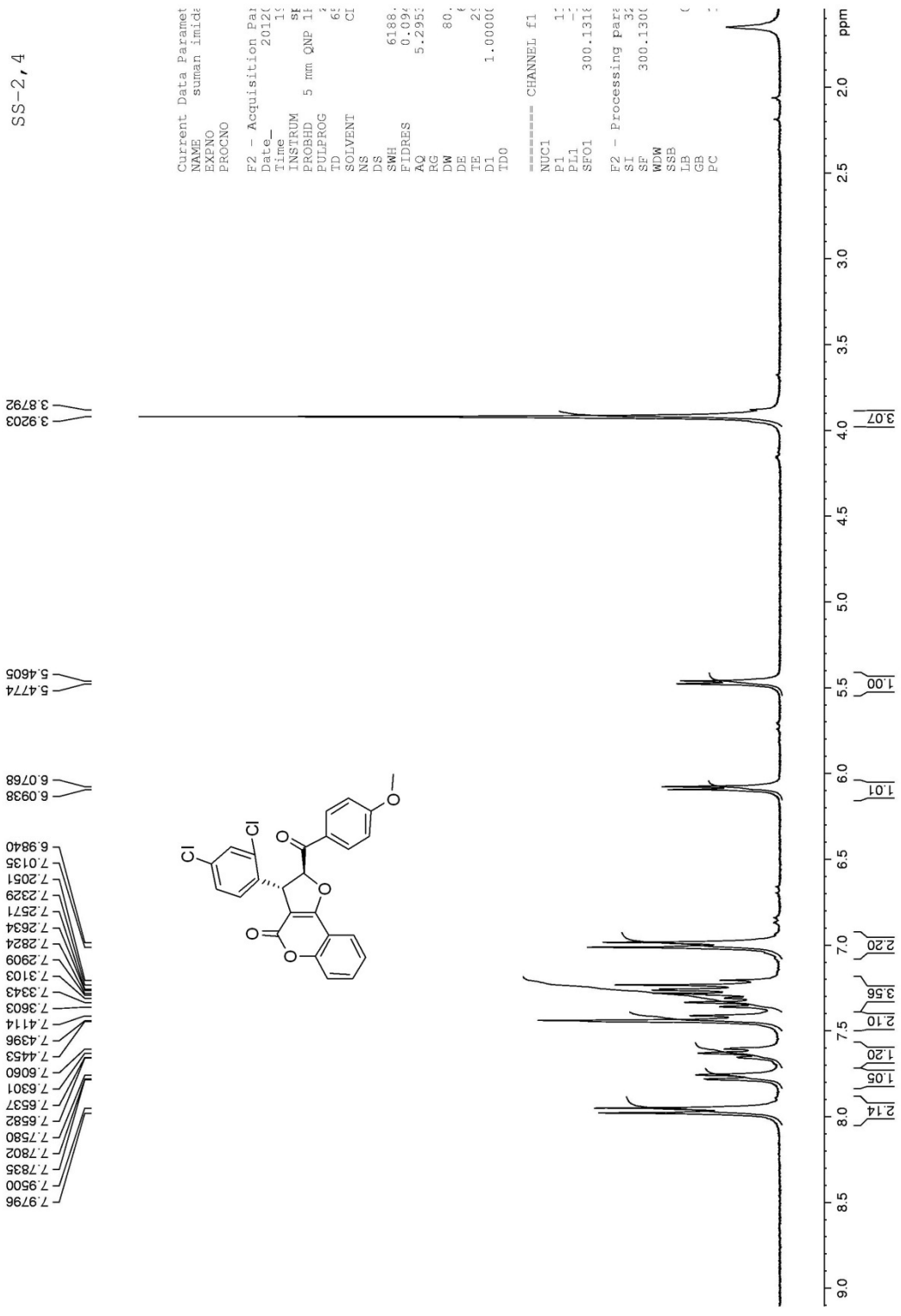


Fig 23: ^{1}H spectra of 3-(2,4-dichlorophenyl)-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

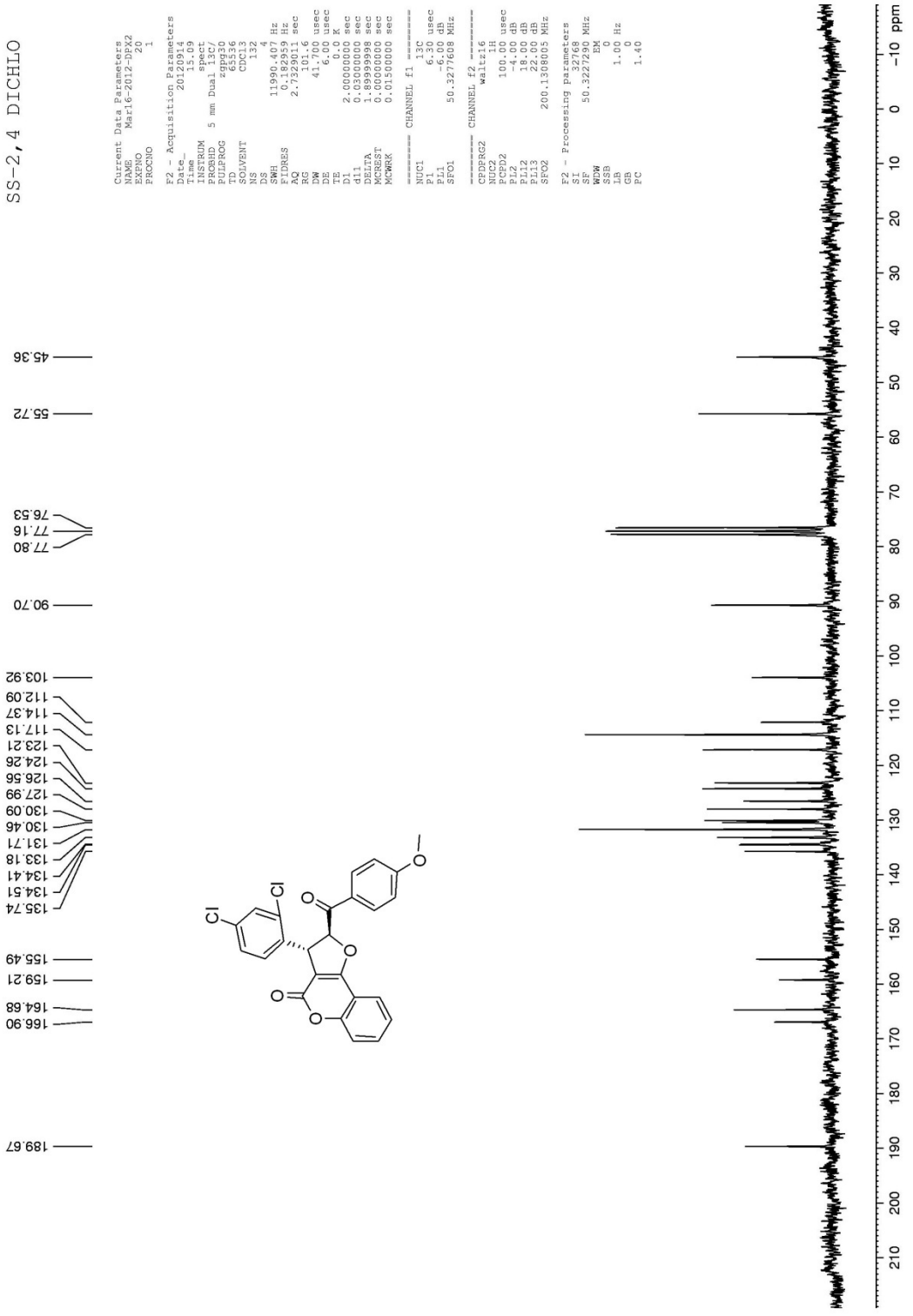


Fig 24: ^{13}C spectra of 3-(2,4-dichlorophenyl)-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one

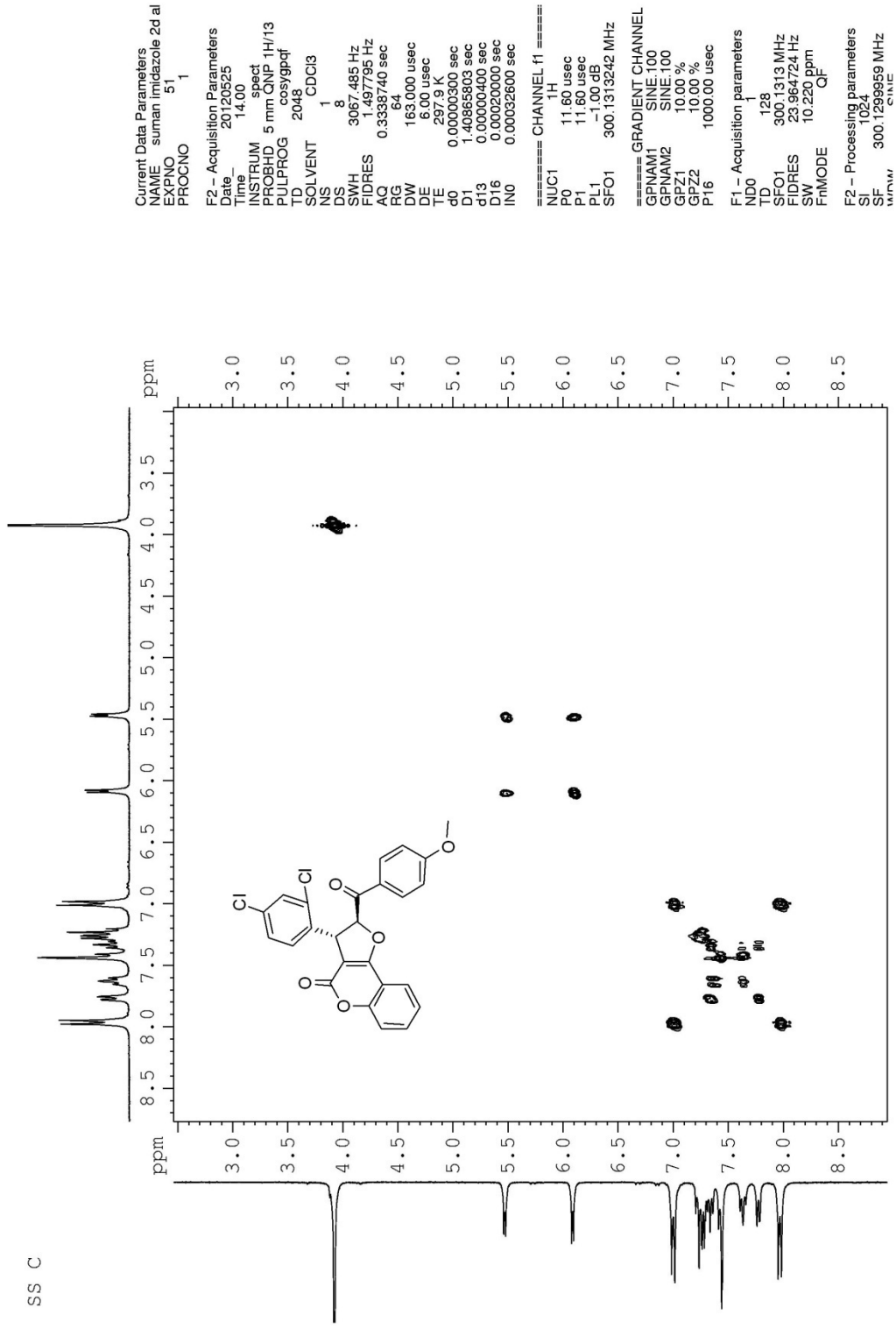


Fig 25: COSY spectra of 3-(2,4-dichlorophenyl)-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

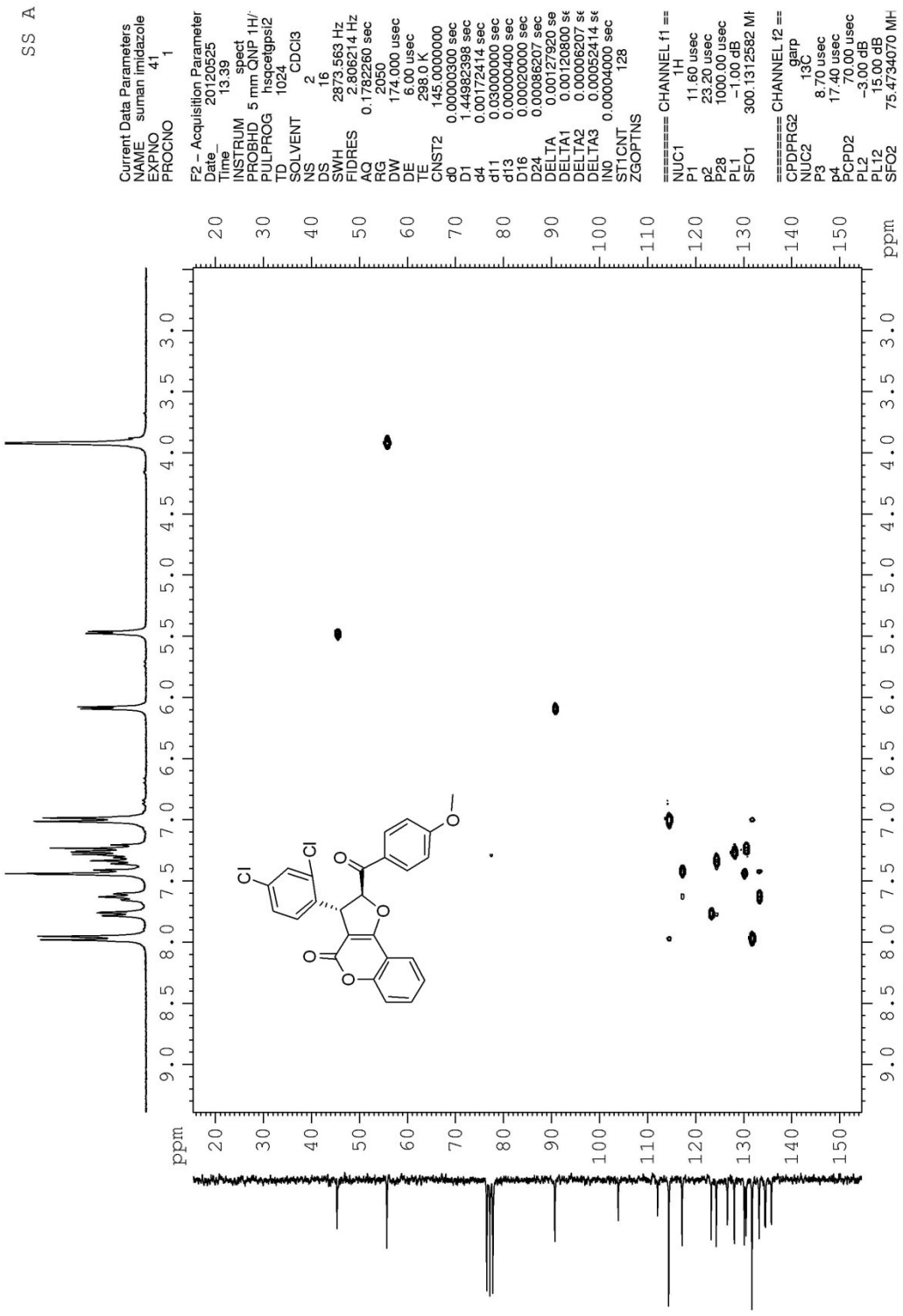


Fig 26: HSQC spectra of 3-(2,4-dichlorophenyl)-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

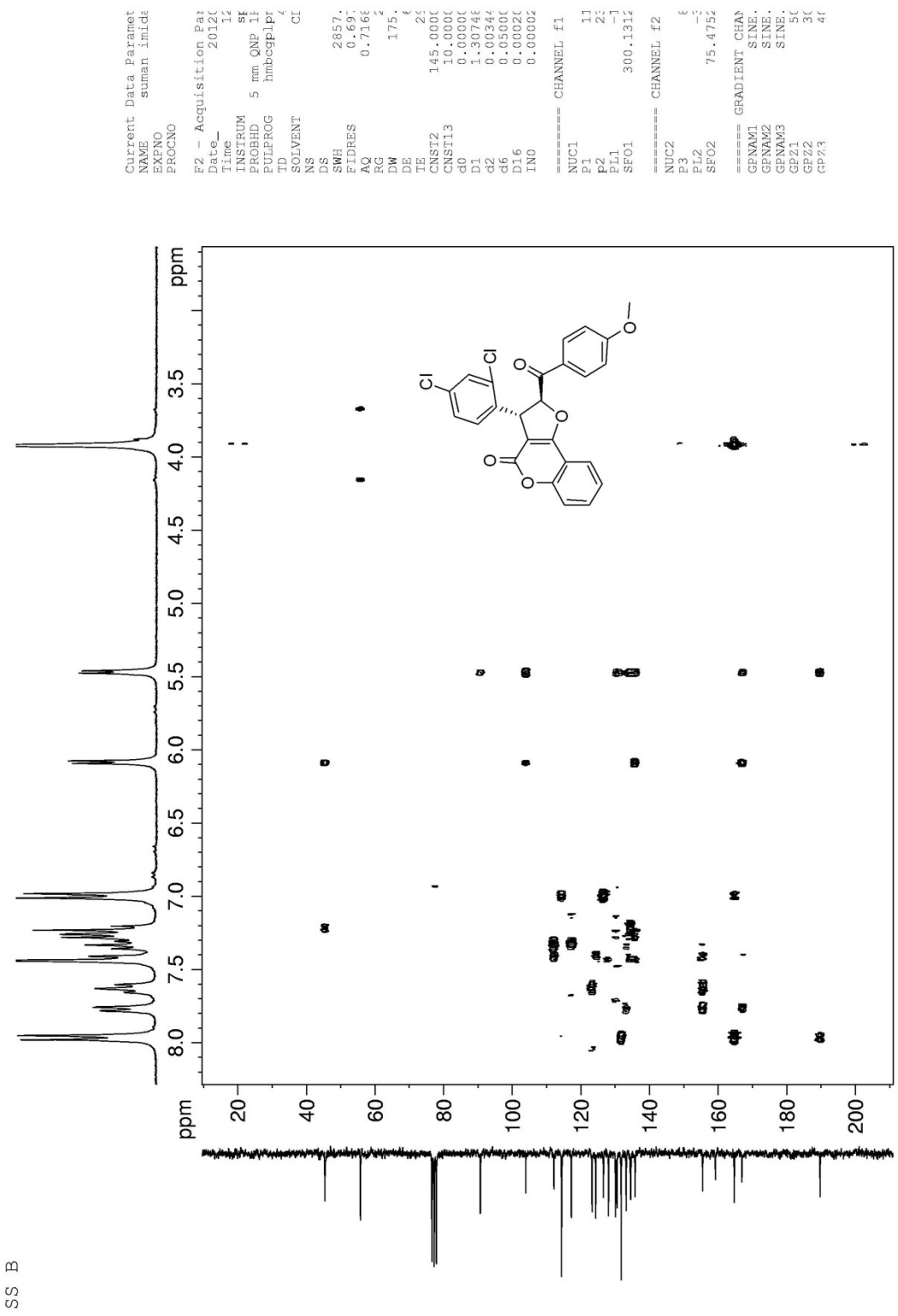


Fig 27: HMBC spectra of 3-(2,4-dichlorophenyl)-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

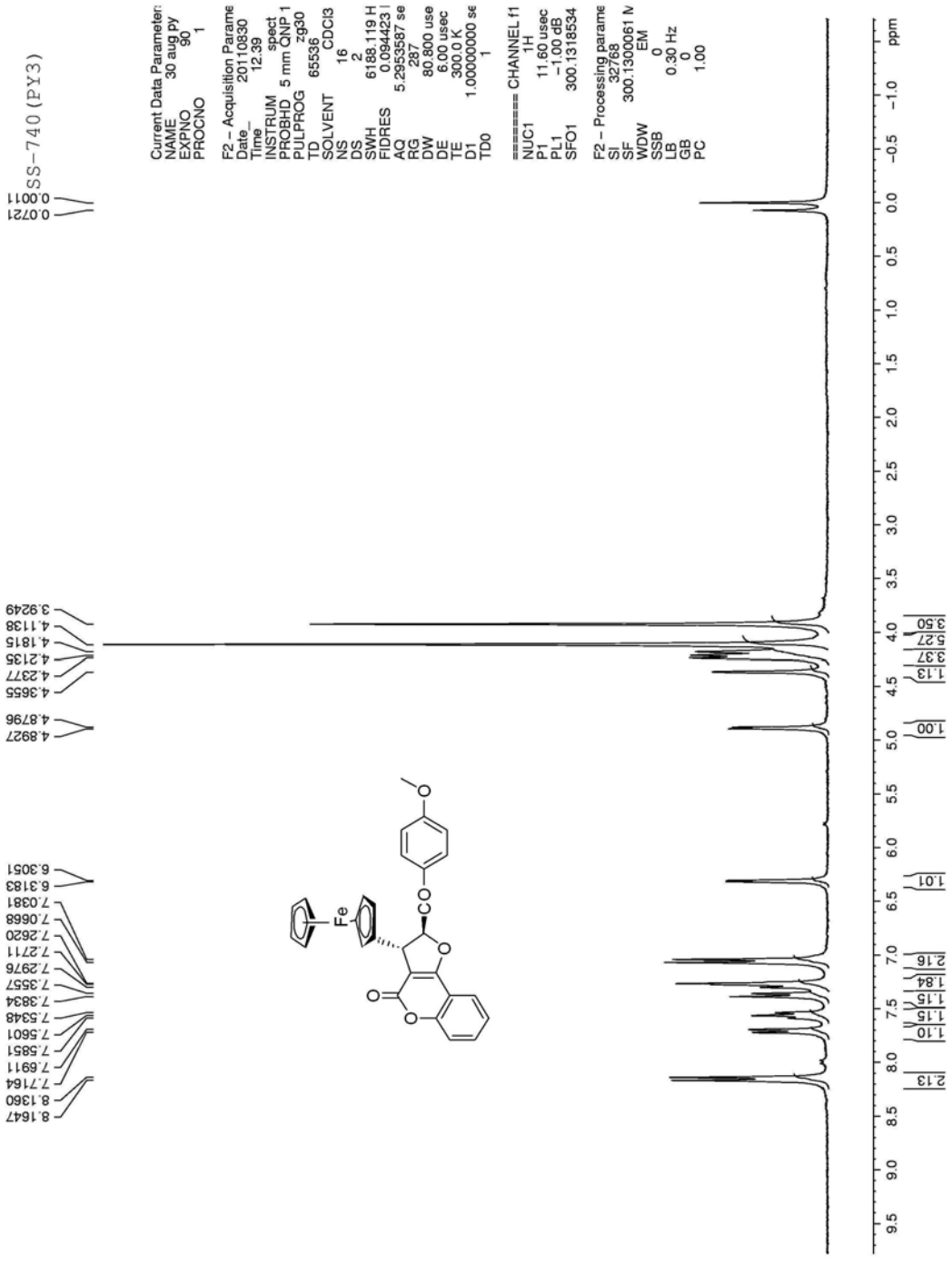


Fig 28: ^{1}H spectra of 3- ferrocenyl-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

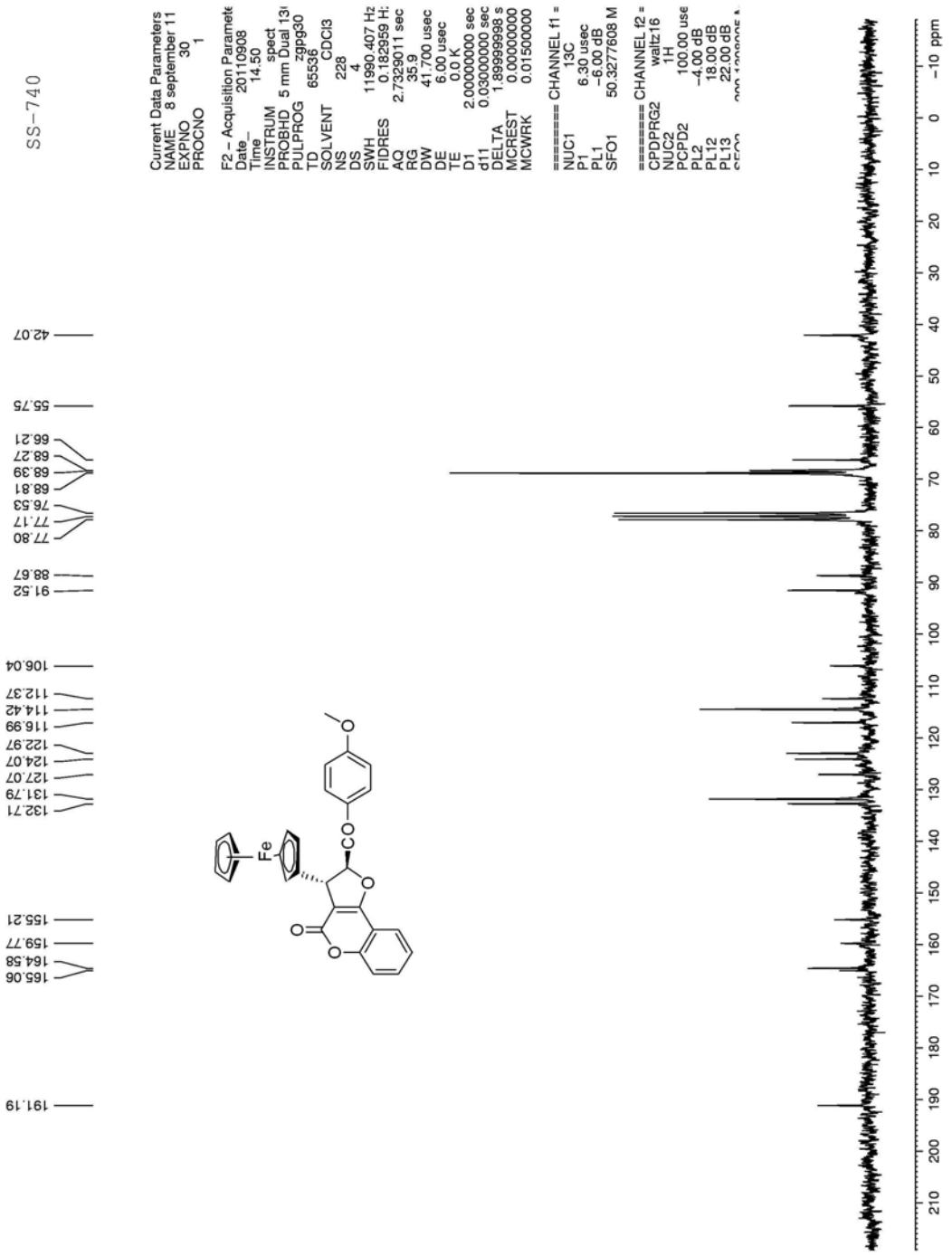


Fig 29: ^{13}C spectra of 3- ferrocenyl-2-(4-methoxybenzoyl)-2H-furo[3,2-c]chromen-4(3H)-one:

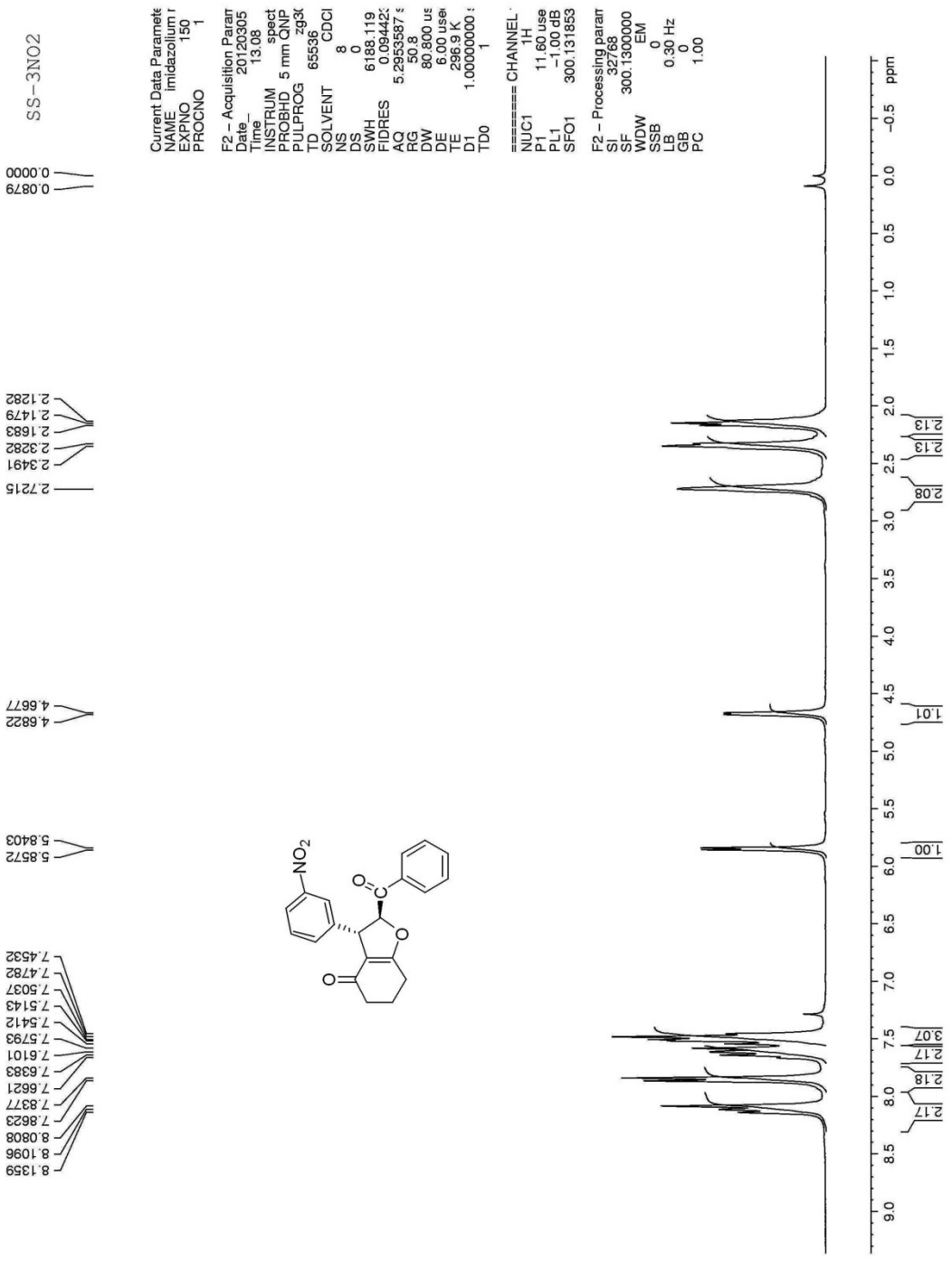


Fig 30: ^{1}H spectra of 2-benzoyl-3-(3-nitrophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

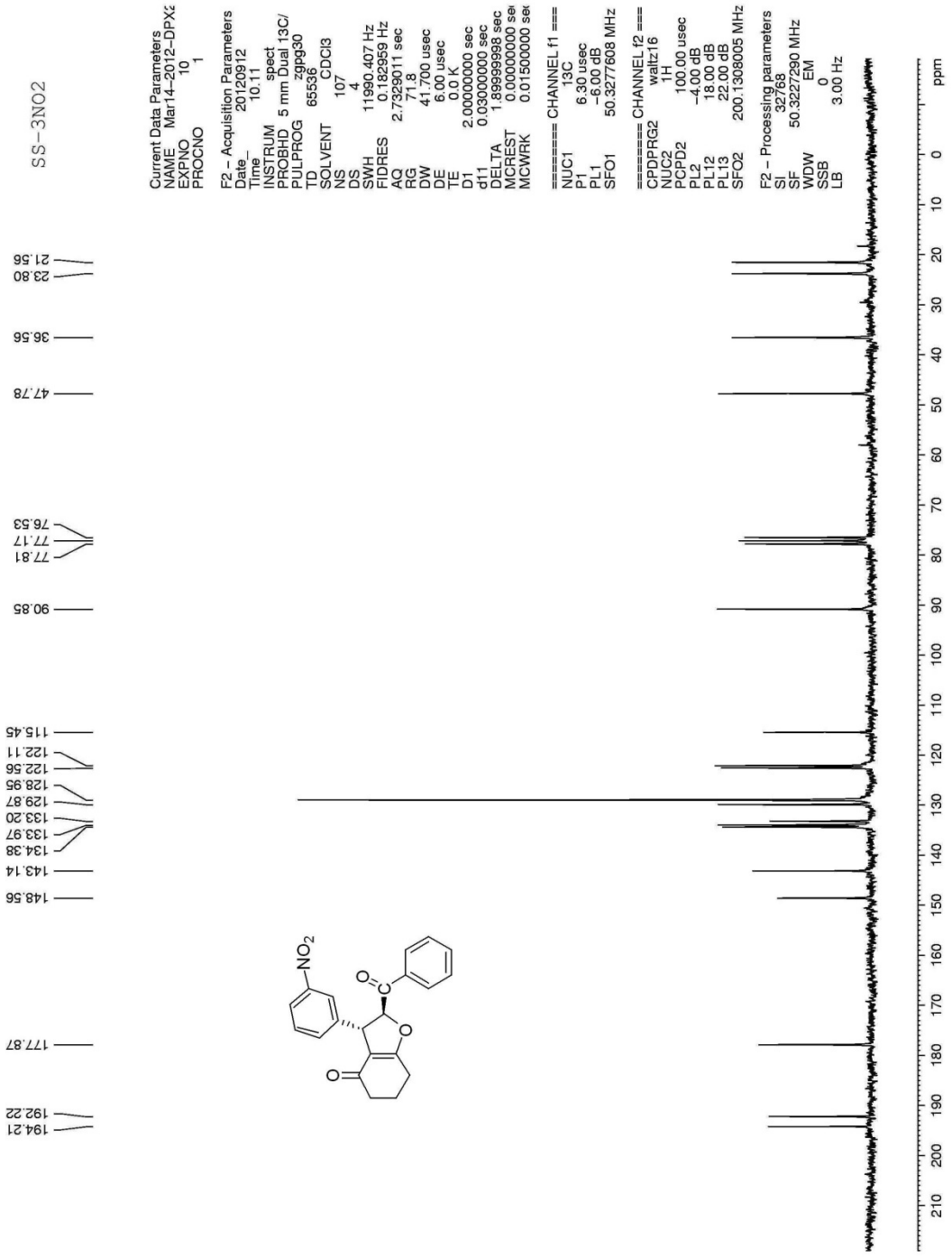


Fig 31: ^{13}C spectra of 2-benzoyl-3-(3-nitrophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

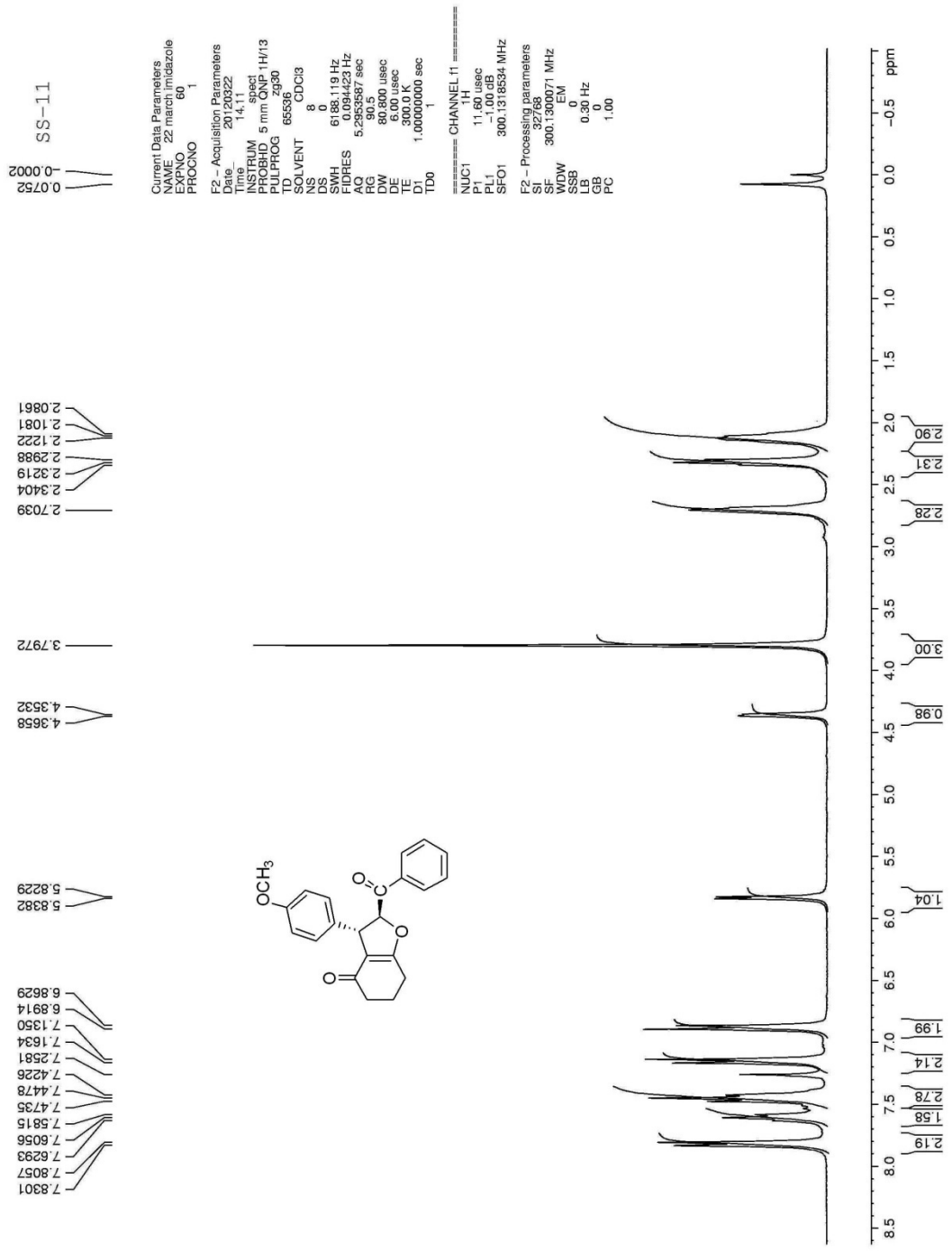


Fig 32. ^{1}H spectra of 2-benzoyl-3-(4-methoxyphenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

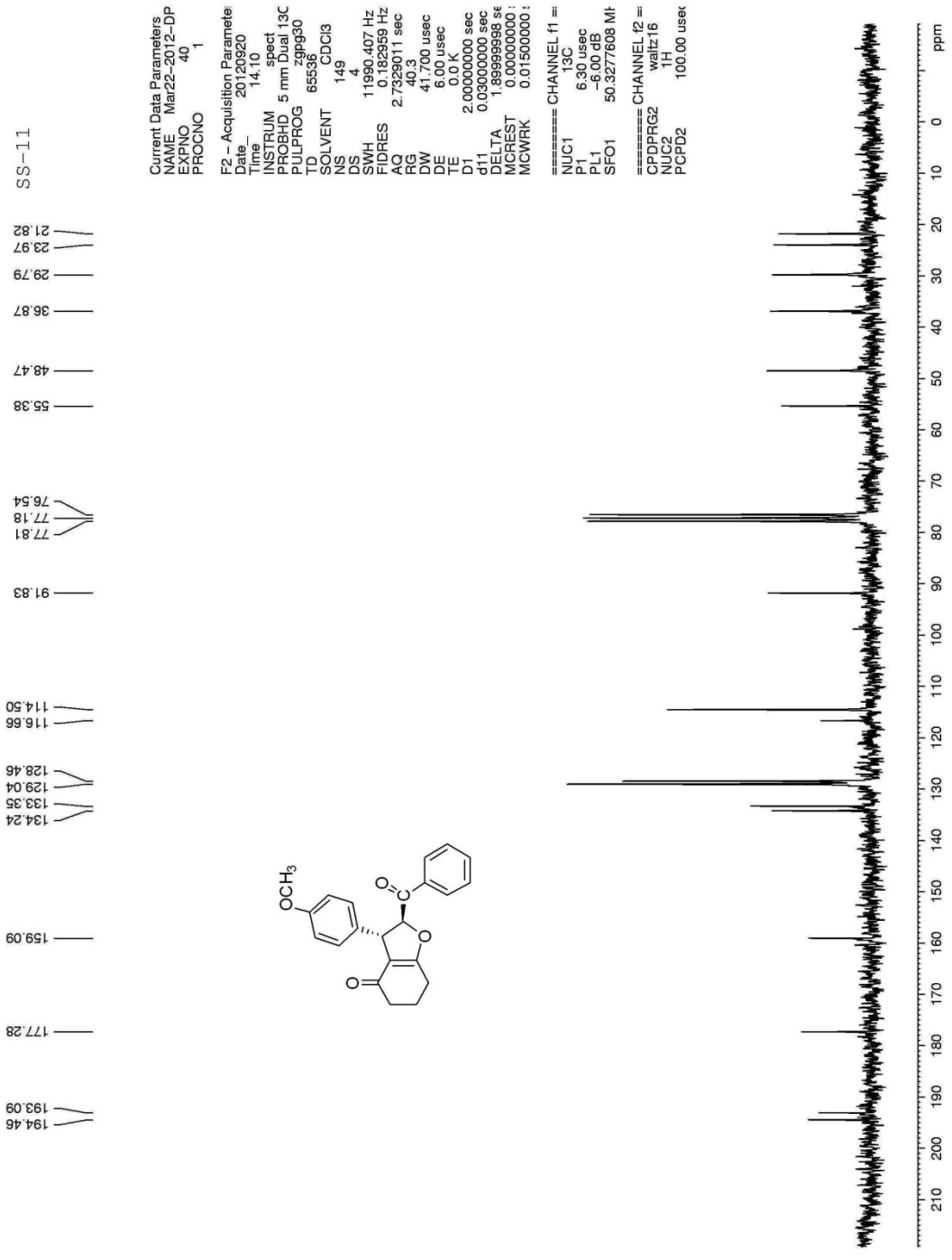


Fig 33: ^{13}C spectra of 2-benzoyl-3-(4-methoxyphenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

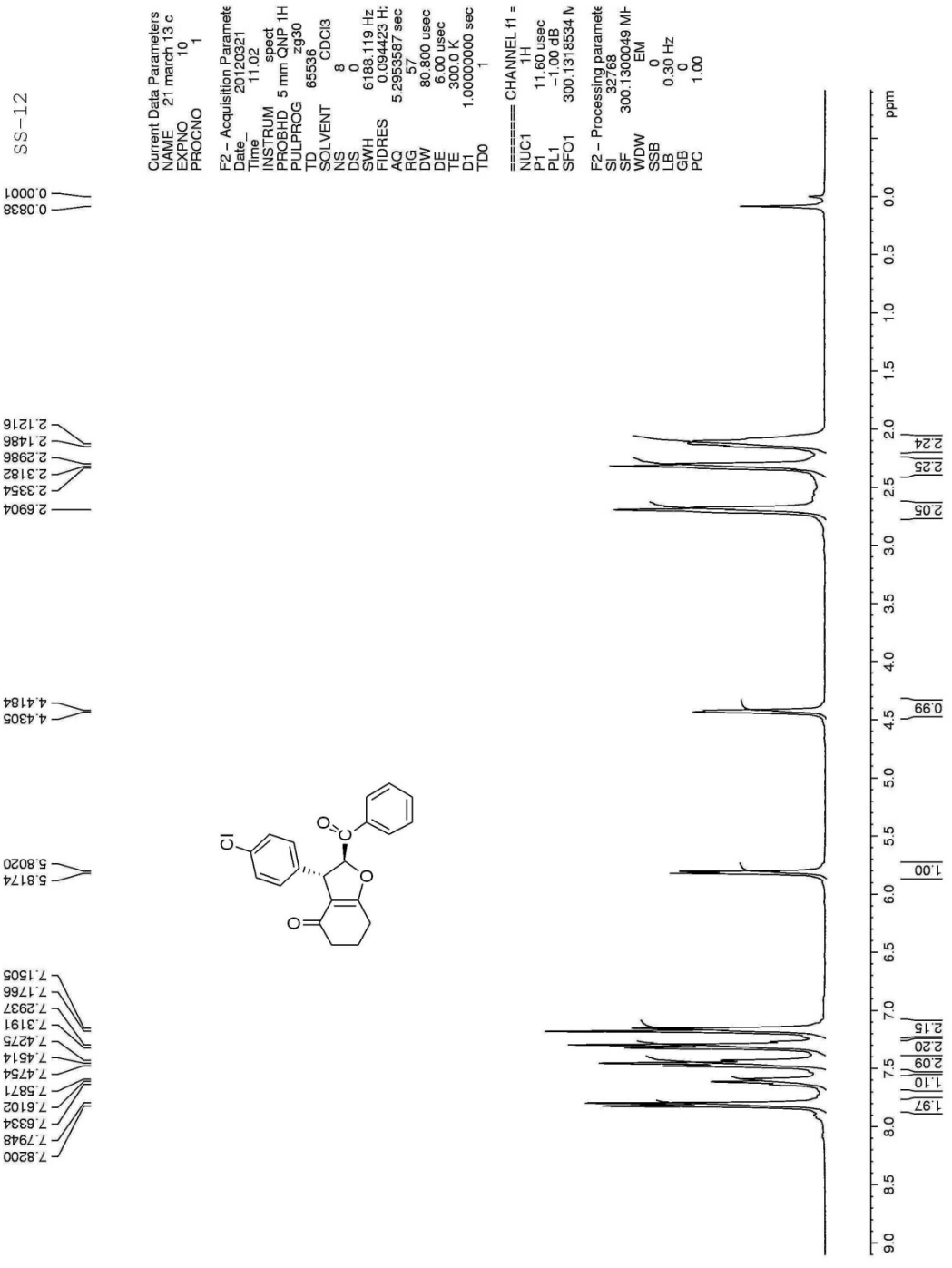


Fig 34: ^{1}H spectra of 2-benzoyl-3-(4-chlorophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

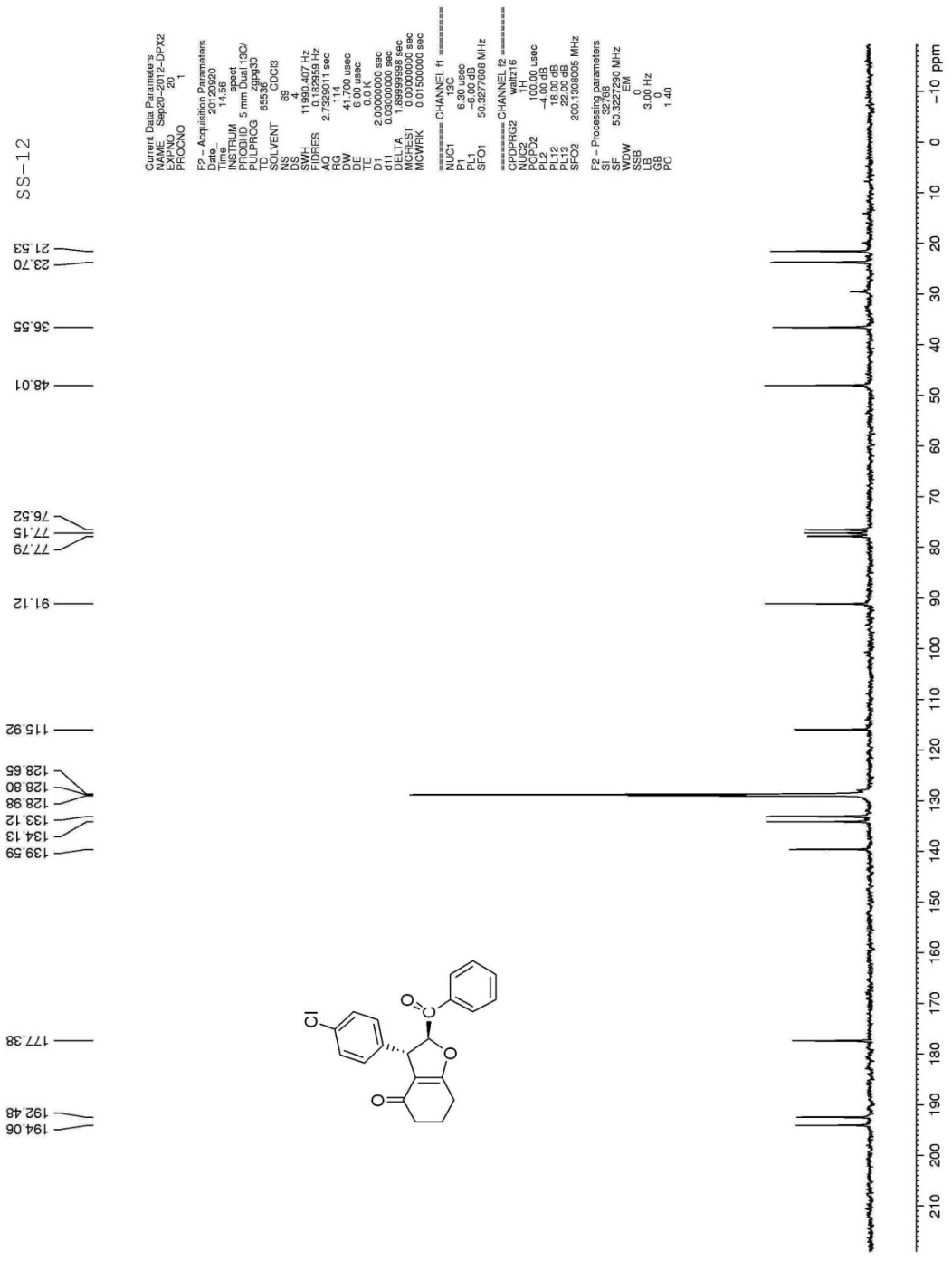


Fig 35: ^{13}C spectra of 2-benzoyl-3-(4-chlorophenyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

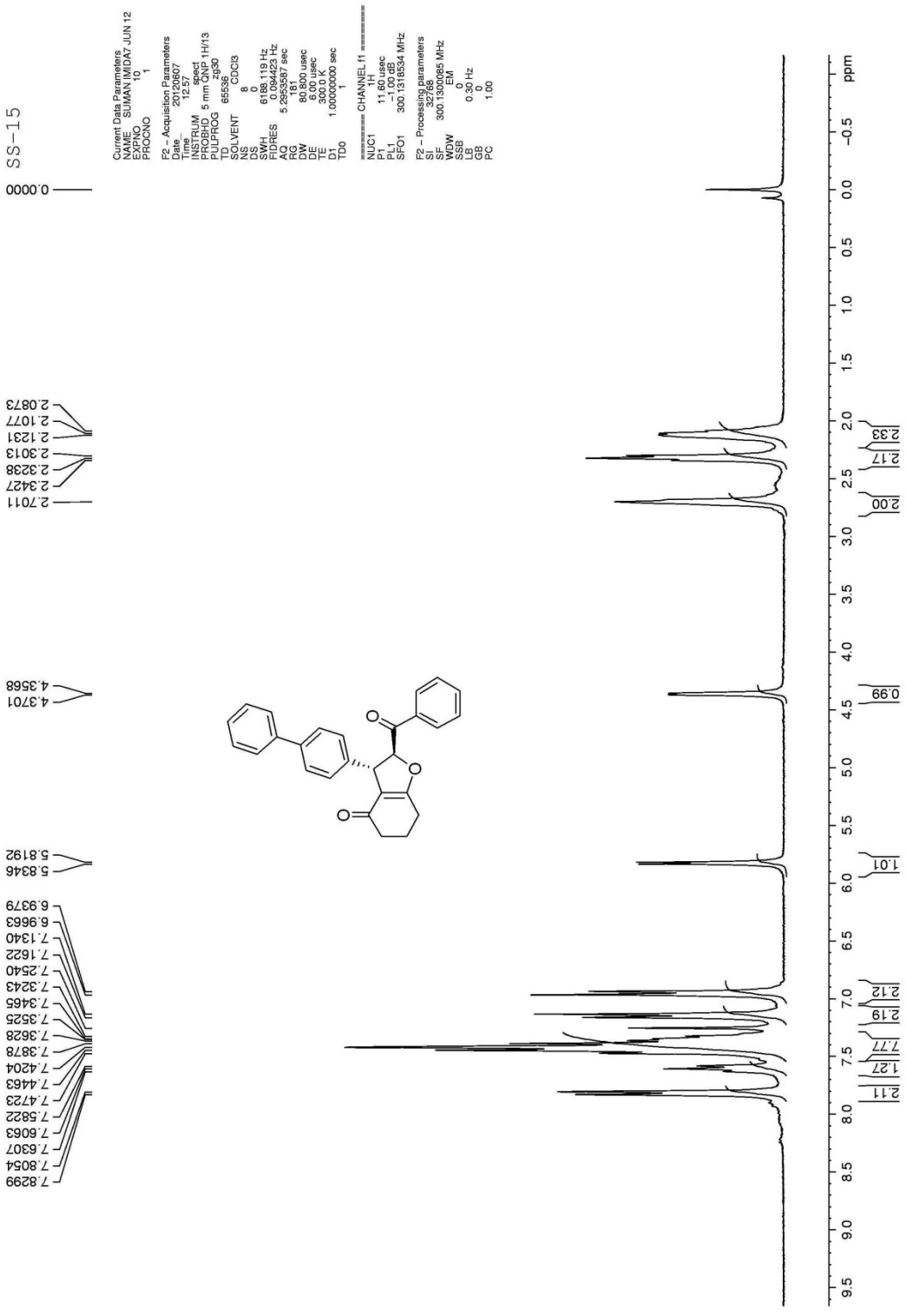


Fig 36: ^{1}H spectra of 2-benzoyl-3-(biphenyl-4-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

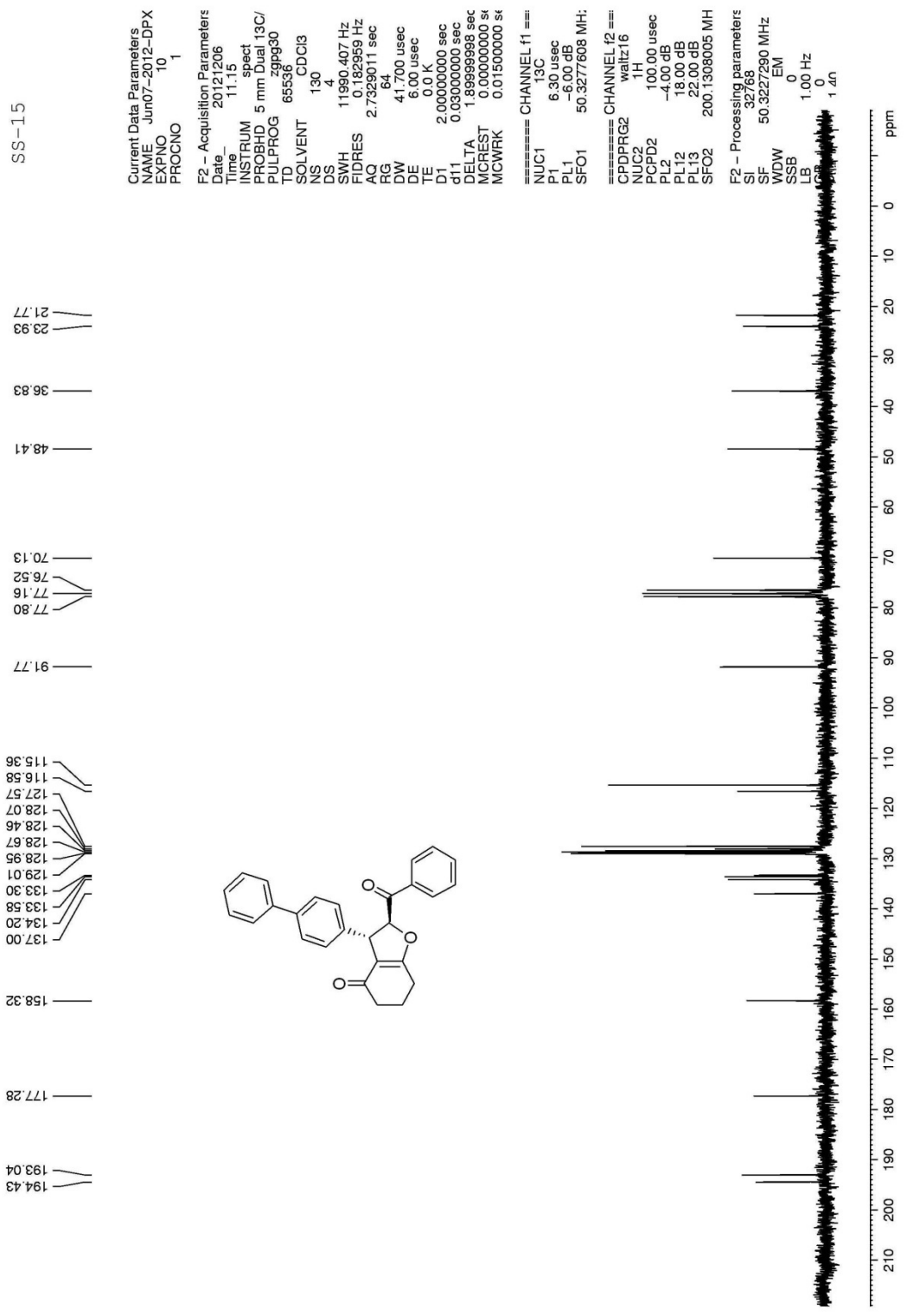


Fig 37: ^{13}C spectra of 2-benzoyl-3-(biphenyl-4-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

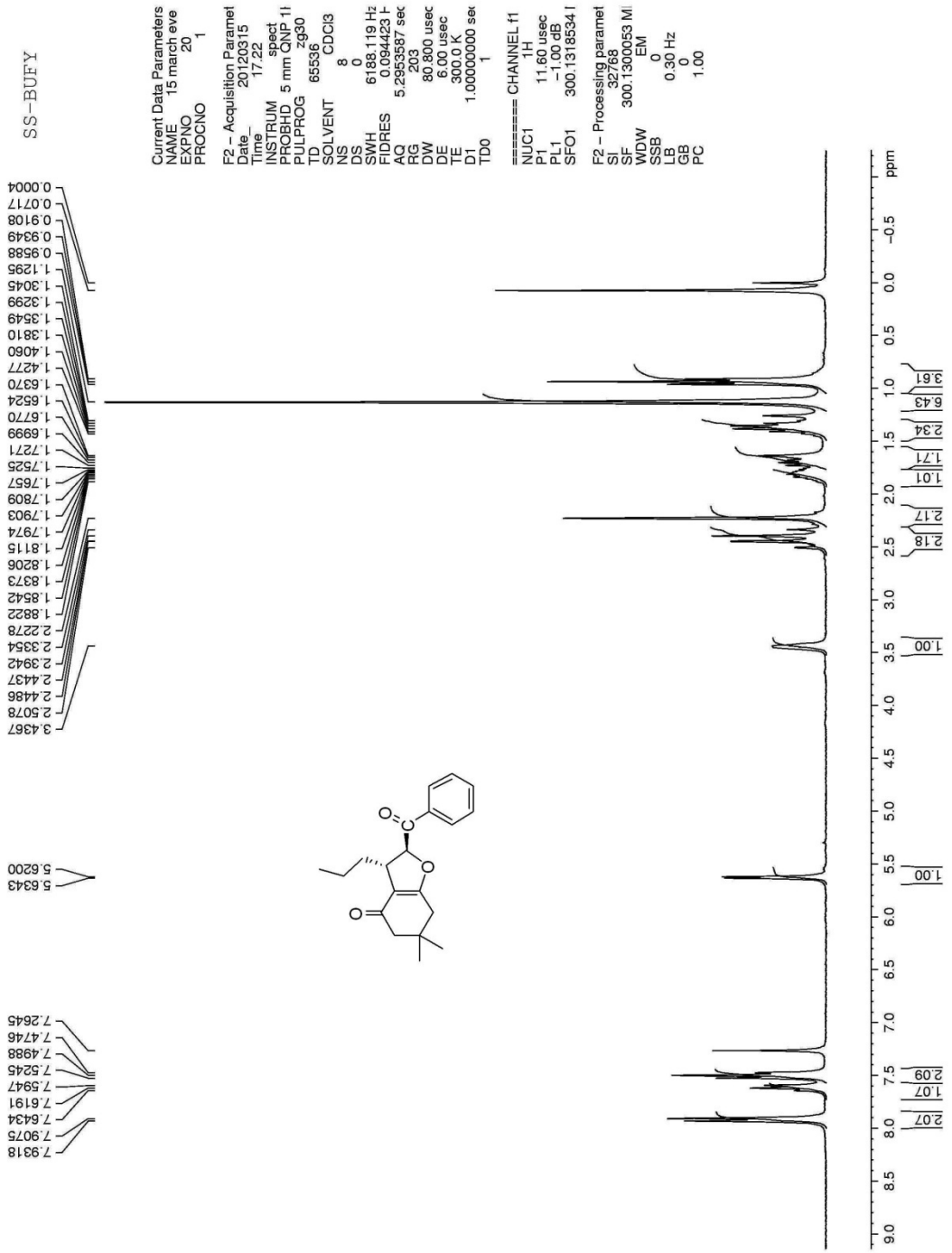


Fig 38: ^{1}H spectra of 2-benzoyl-6,6-dimethyl-3-propyl-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

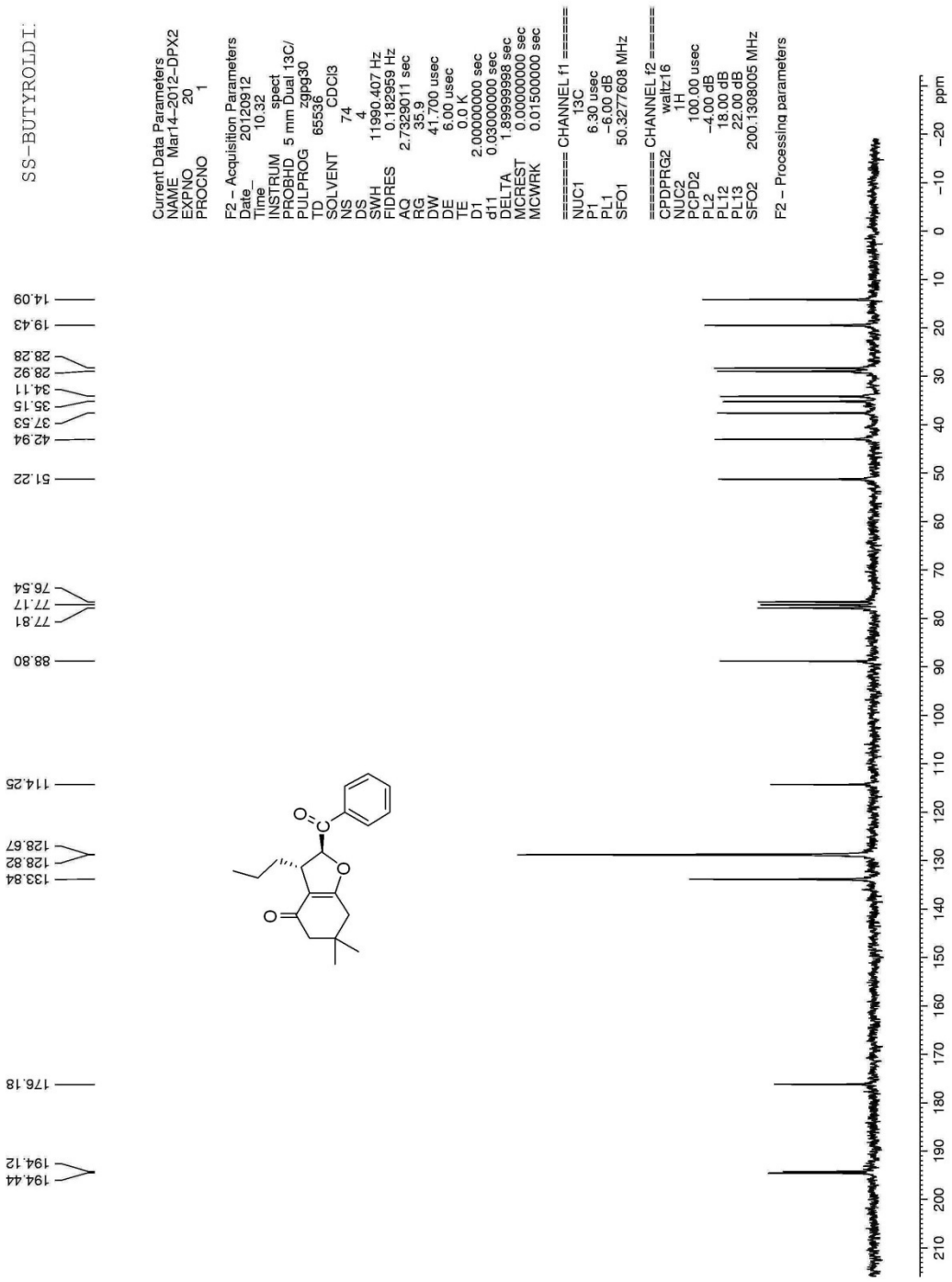


Fig 39: ^{13}C spectra of 2-benzoyl-6,6-dimethyl-3-propyl-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

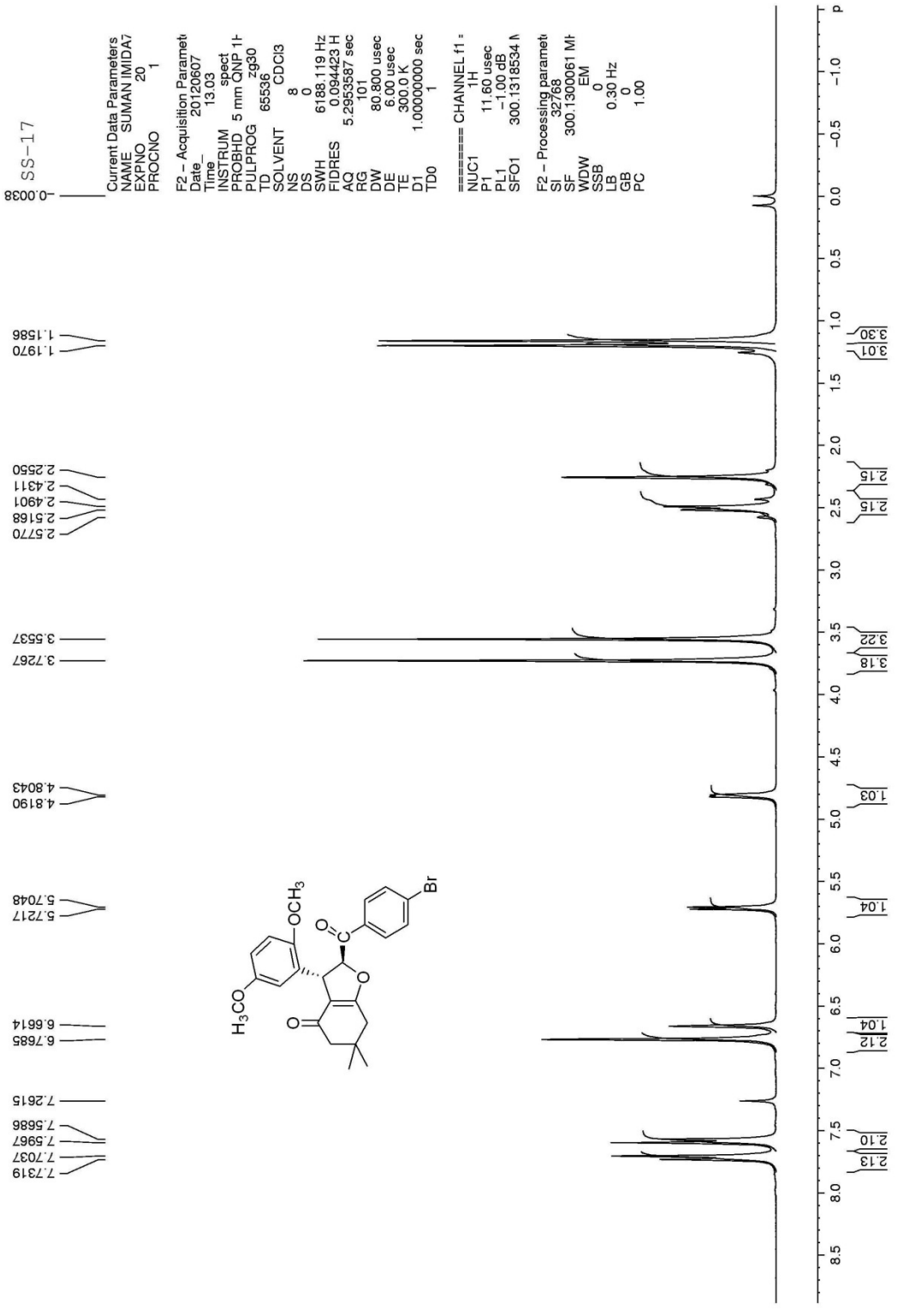


Fig 40: ^{1}H spectra of 2-(4-bromobenzoyl)-3-(2,5-dimethoxyphenyl)-6,6-dimethyl-2,3,6,7-tetrahydrobenzofuran-4(5H)-one :

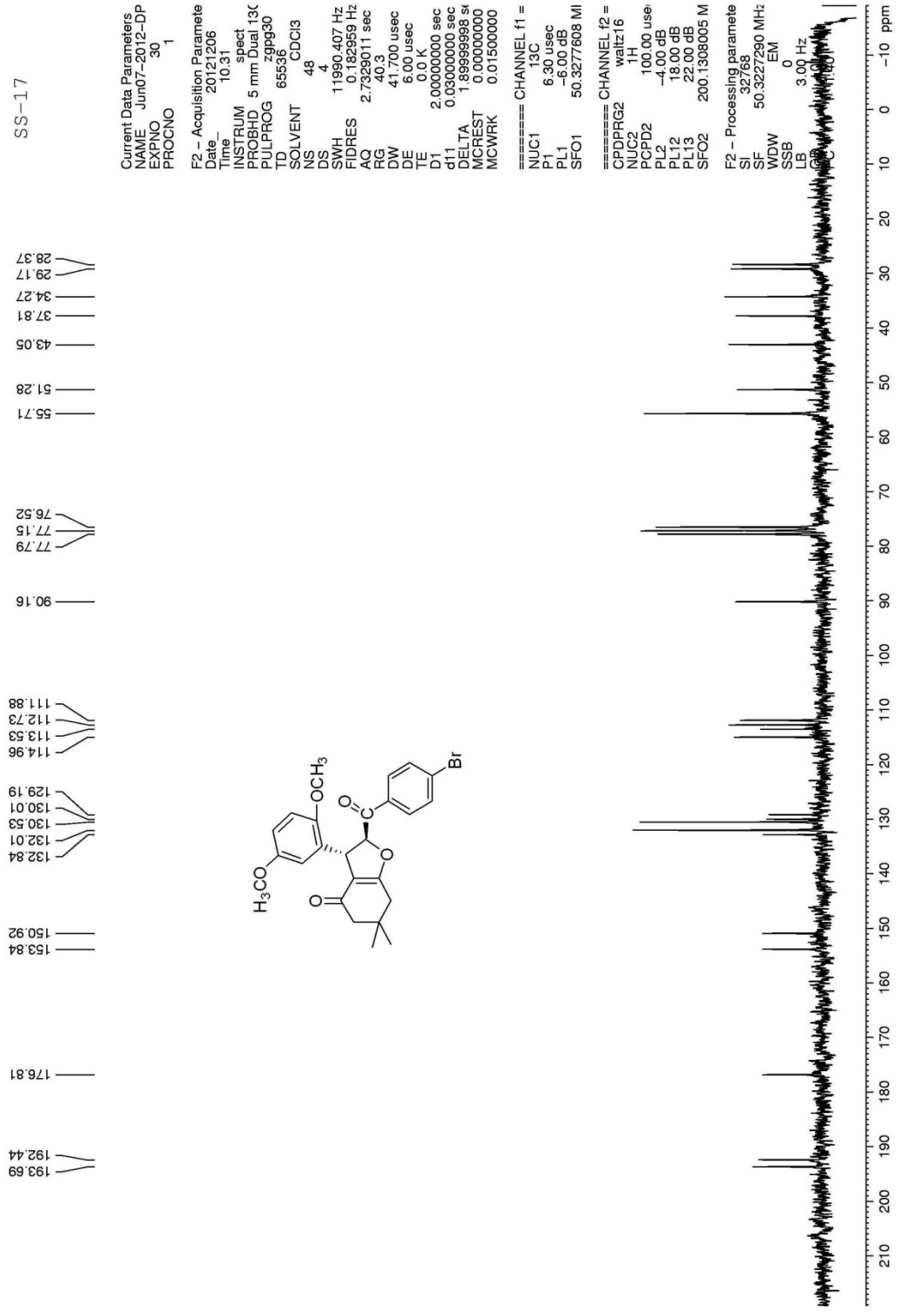


Fig 41: ^{13}C spectra of 2-(4-bromobenzoyl)-3-(2,5-dimethoxyphenyl)-6,6-dimethyl-2,3,6,7-tetrahydrobenzofuran-4(5H)-one :

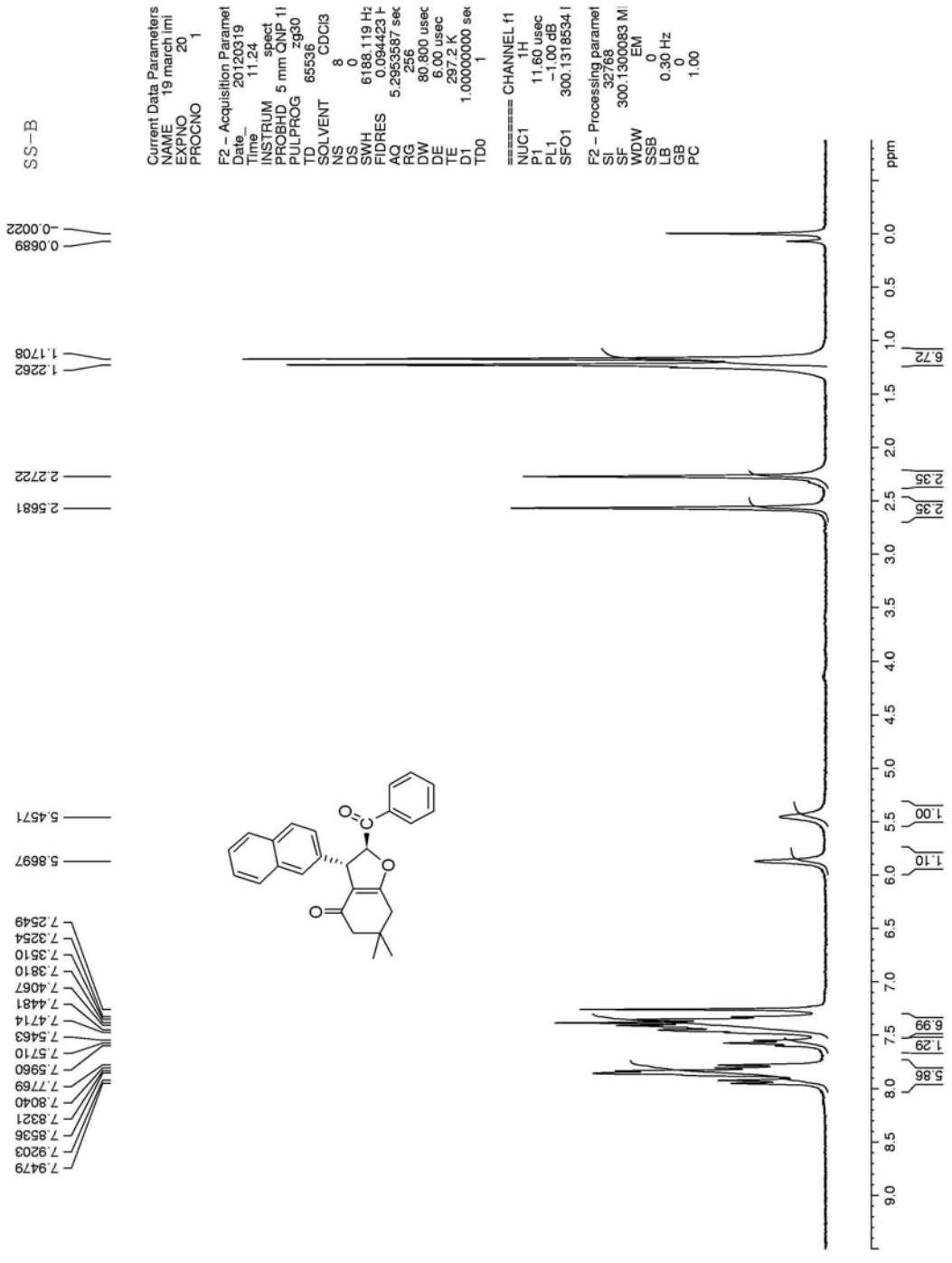


Fig 42: ^{1}H spectra of 2-benzoyl-6,6-dimethyl-3-(naphthalen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

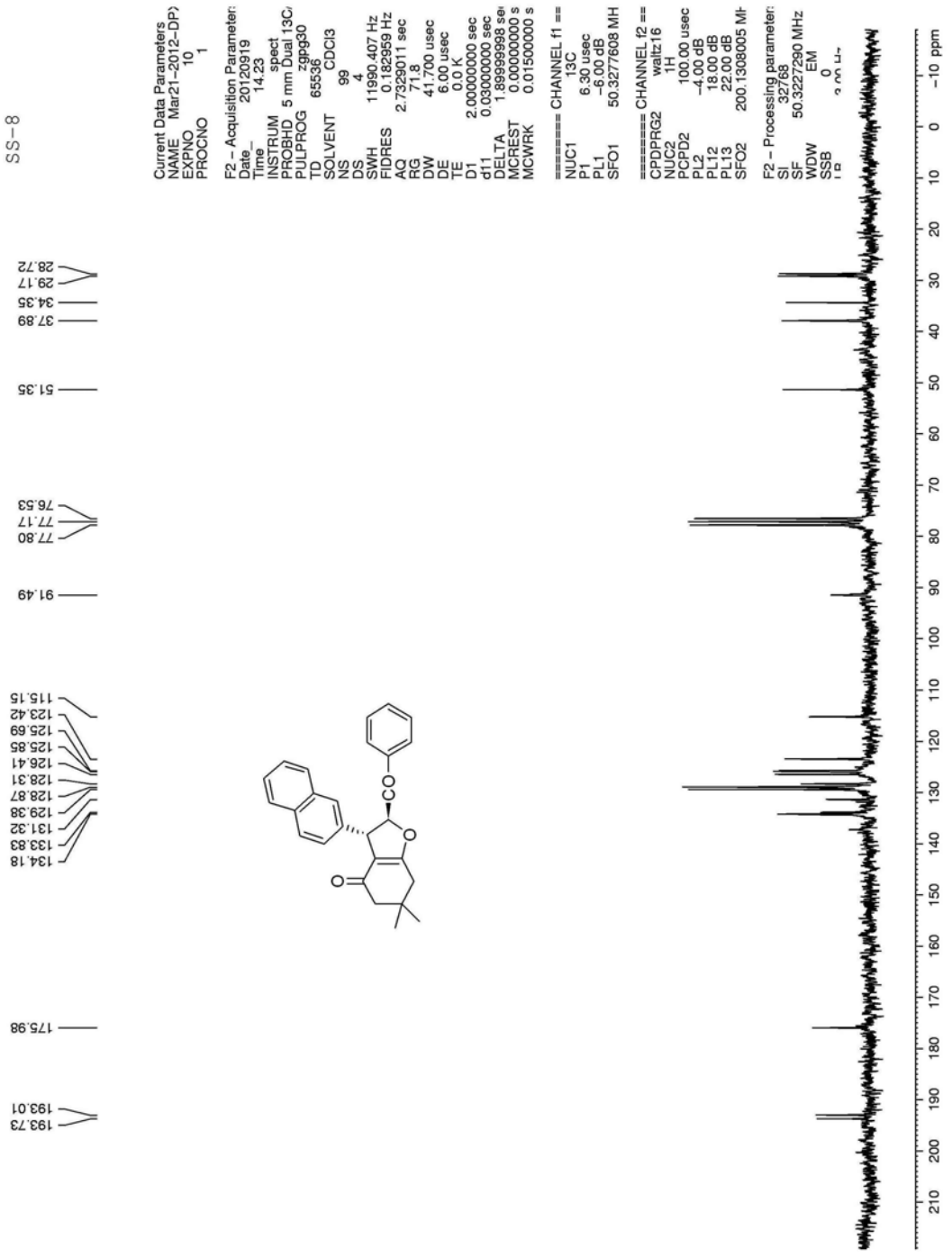


Fig 43: ^{13}C spectra of 2-benzoyl-6,6-dimethyl-3-(naphthalen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

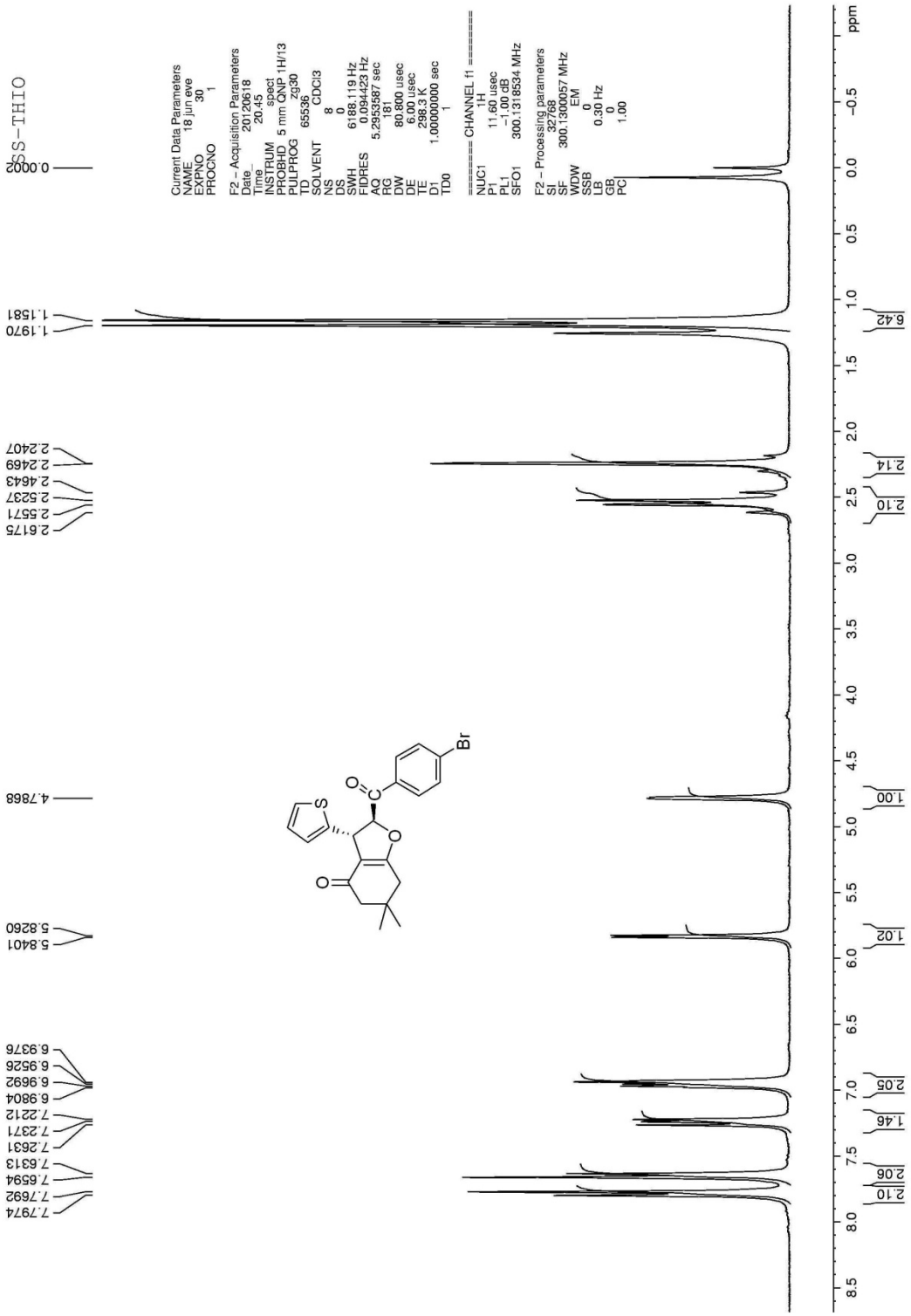


Fig 44: ^{1}H spectra of 2-(4-bromobenzoyl)-6,6-dimethyl-3-(thiophen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

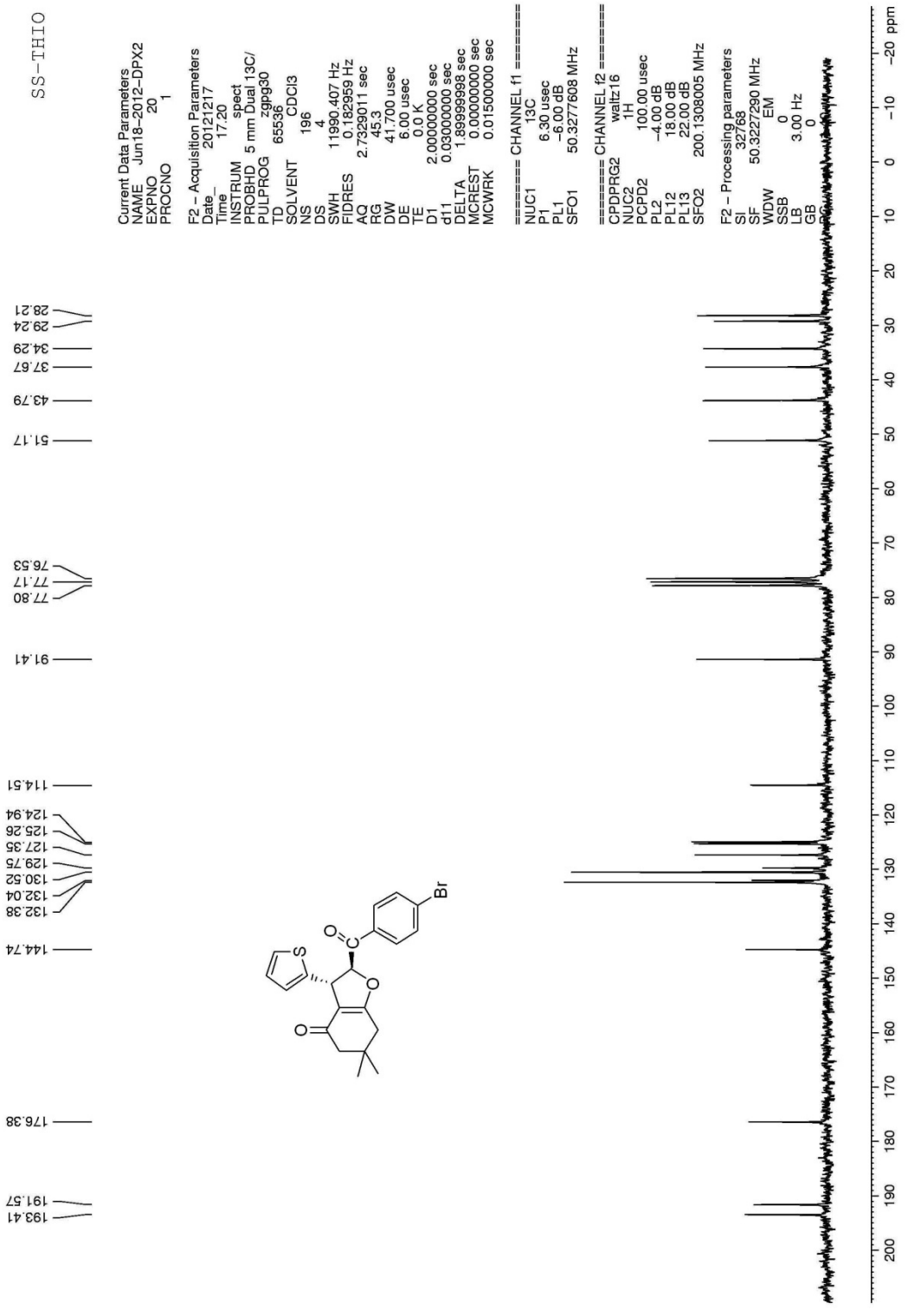


Fig 45: ^{13}C spectra of 2-(4-bromobenzoyl)-6,6-dimethyl-3-(thiophen-2-yl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

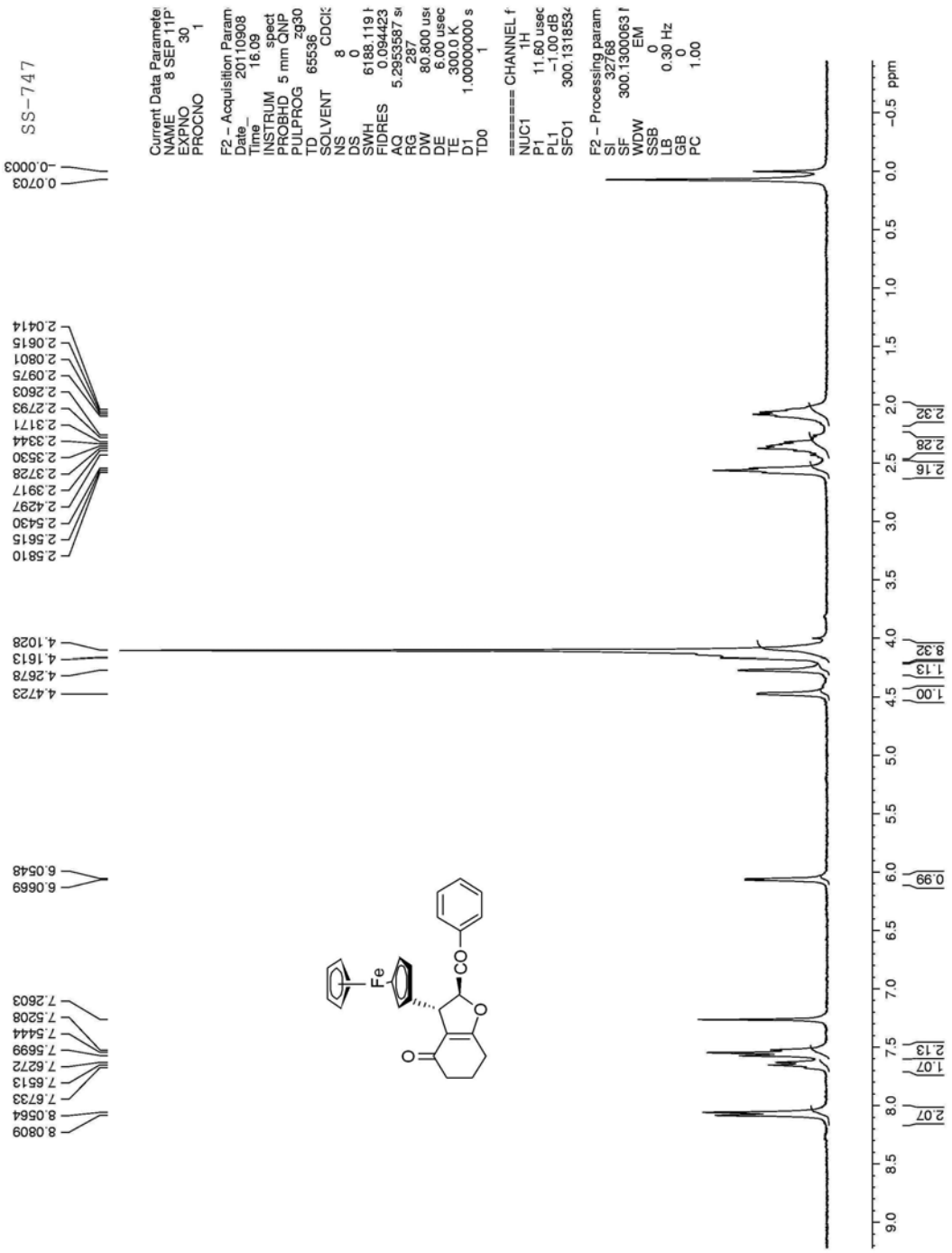


Fig 46: ^{1}H spectra of 2-benzoyl-3- ferrocenyl -2, 3, 6, 7-tetrahydrobenzofuran-4(5H)-one:

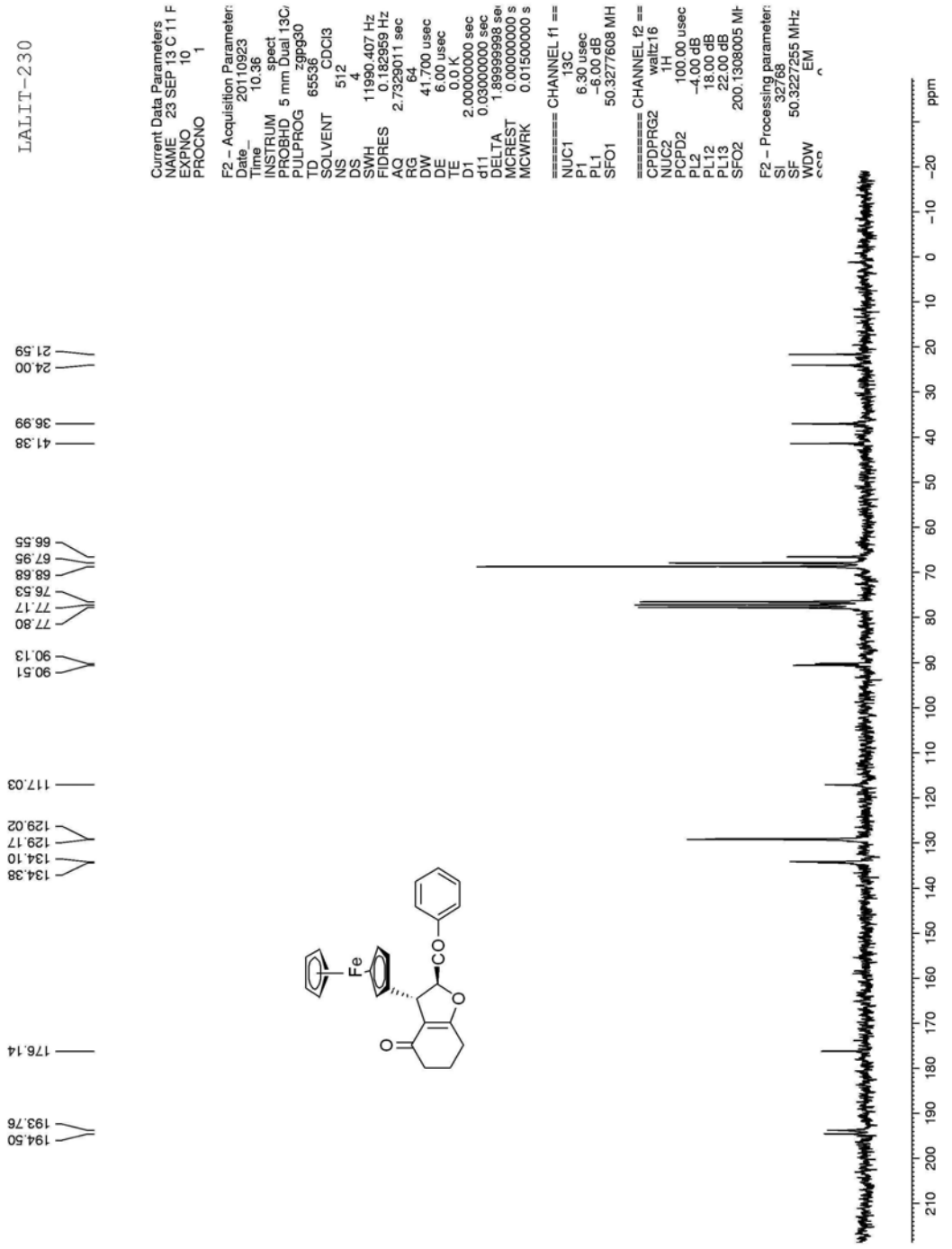


Fig 47: ^{13}C spectra of 2-benzoyl-3- ferrocenyl -2, 3, 6, 7-tetrahydrobenzofuran-4(5H)-one :

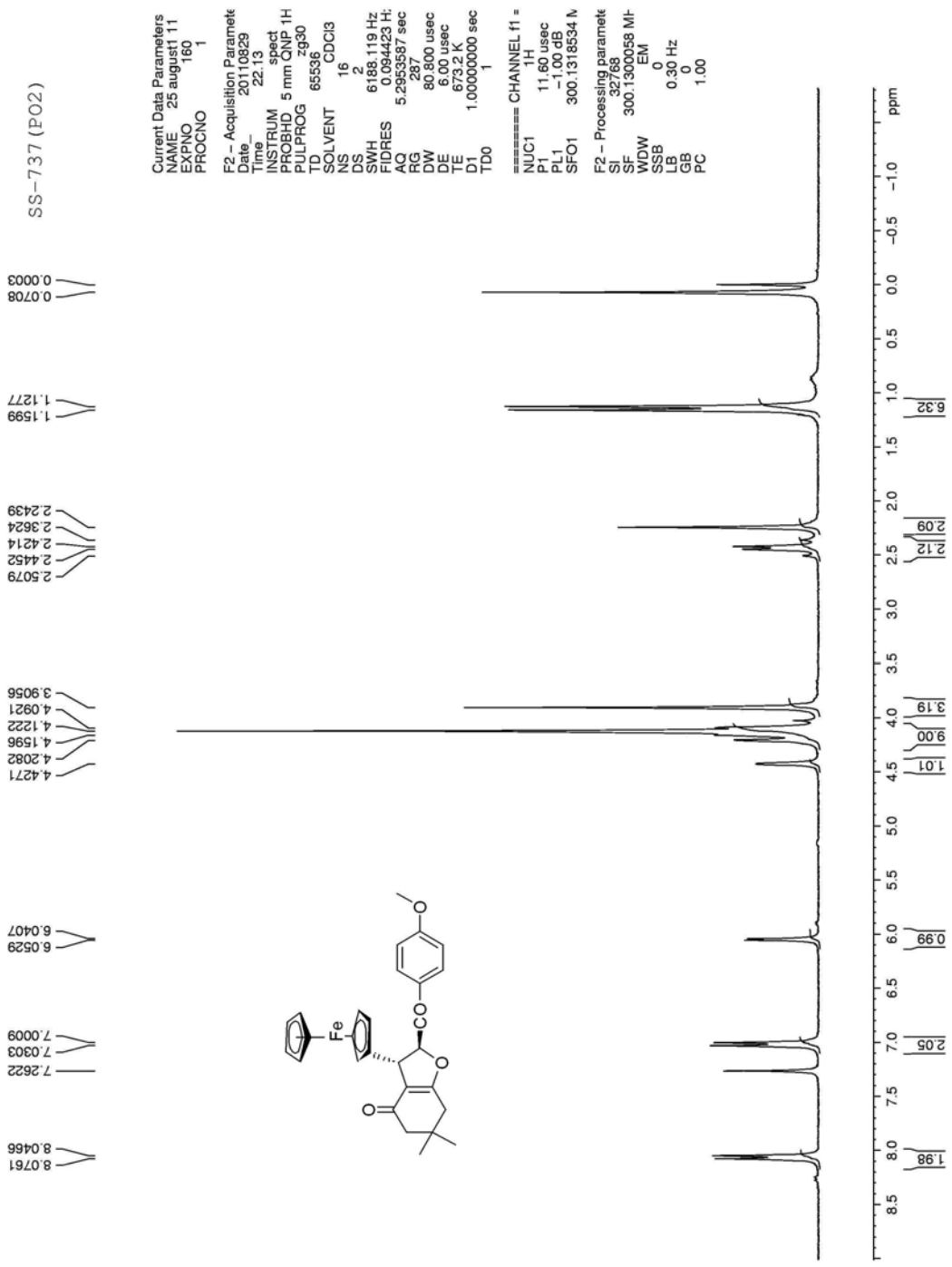


Fig 48: ^{1}H spectra of 6,6-dimethyl-3-ferrocenyl-2-(4-methoxybenzoyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one:

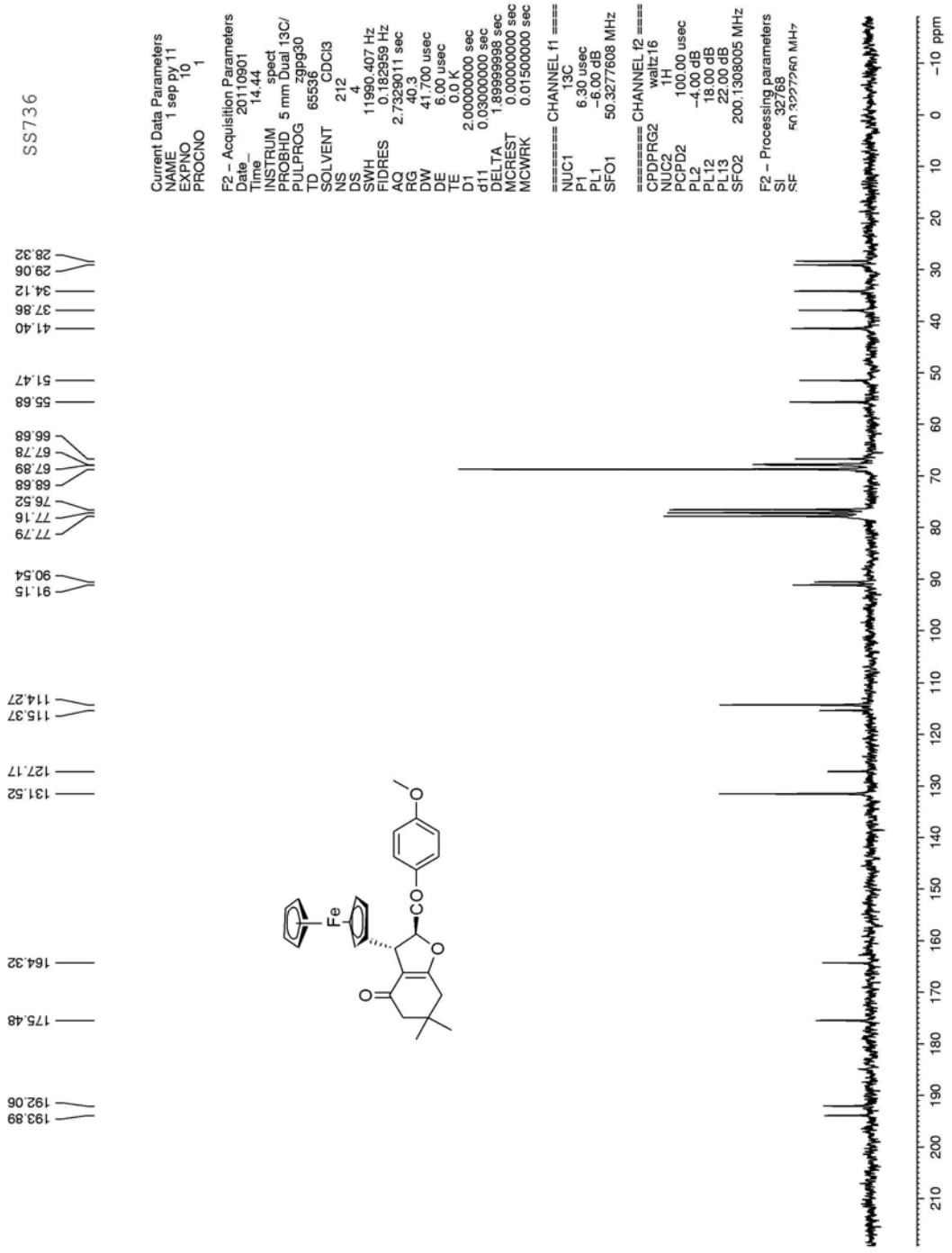


Fig 49: ^{13}C spectra of 6, 6-dimethyl, 3-ferrocenyl 2-(4-methoxybenzoyl)-2,3,6,7-tetrahydrobenzofuran-4(5H)-one: