

Regioselective synthesis of unsymmetrical biheteroaryls via copper(II)-catalyzed cascade annulation

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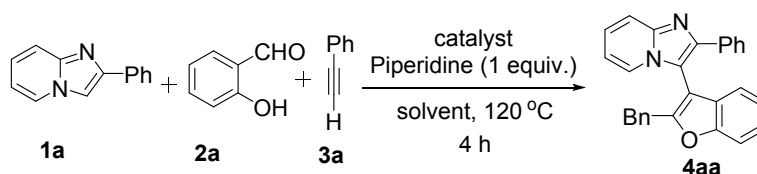
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1. General information:

All reagents were purchased from commercial sources and used without further purification. ^1H NMR spectra were determined on 400 MHz spectrometer as solutions in CDCl_3 . Chemical shifts are expressed in parts per million (δ) and the signals were reported as s (singlet), d (doublet), t (triplet), m (multiplet) and coupling constants (J) were given in Hz. $^{13}\text{C}\{^1\text{H}\}$ NMR spectra were recorded at 100 MHz in CDCl_3 solution. Chemical shifts as internal standard are referenced to CDCl_3 ($\delta = 7.26$ for ^1H and $\delta = 77.16$ for $^{13}\text{C}\{^1\text{H}\}$ NMR) as internal standard. TLC was done on silica gel coated glass slide. All solvents were dried and distilled before use. Commercially available solvents were freshly distilled before the reaction. All reactions involving moisture sensitive reactants were executed using oven dried glassware. All the imidazoheterocycles were prepared by our reported method.^{1a,b}

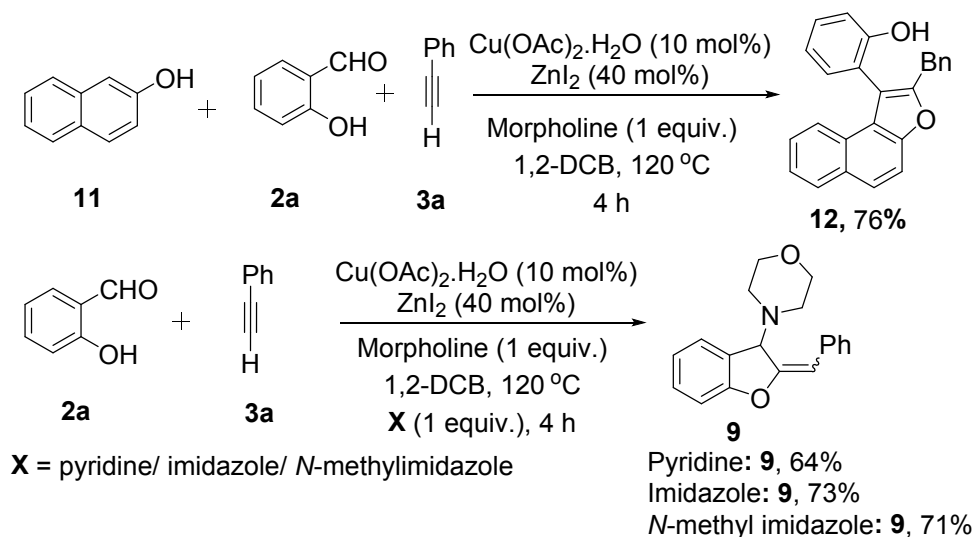
2. Optimization table^a



Entry	Catalyst (mol%)	Additives (mol%)	Solvent	Yields (%)
1	CuI (10)	ZnI ₂ (20)	CH ₃ CN	57
2	CuI (10)	ZnI ₂ (20)	Toluene	26
3	CuI (10)	ZnI ₂ (20)	1,4-dioxane	11
4	CuI (10)	ZnI ₂ (20)	1,2-DCE	28
5	CuI (10)	ZnI ₂ (20)	Chlorobenzene	15
6	CuI (10)	ZnI ₂ (20)	1,2-DCB	72
7	CuI (10)	ZnI ₂ (20)	DMF	56
8	Cu ₂ O (10)	ZnI ₂ (20)	1,2-DCB	42
9	CuBr (10)	ZnI ₂ (20)	1,2-DCB	61
10	Cu(OAc) ₂ ·H ₂ O (10)	ZnI ₂ (20)	1,2-DCB	80
11	Cu(OTf) ₂ (10)	ZnI ₂ (20)	1,2-DCB	79
12	CuBr ₂ (10)	ZnI ₂ (20)	1,2-DCB	82
13	CuCl ₂ (10)	ZnI ₂ (20)	1,2-DCB	78
14	Cu(OAc) ₂ ·H ₂ O (10)	AgOTf (20)	1,2-DCB	43
15	Cu(OAc) ₂ ·H ₂ O (10)	1,10-phenanthroline (20)	1,2-DCB	26
16	Cu(OAc) ₂ ·H ₂ O (10)	Zn(OTf) ₂ (20)	1,2-DCB	35
17	Cu(OAc) ₂ ·H ₂ O (10)	-	1,2-DCB	49
18	-	ZnI ₂ (20)	1,2-DCB	16
19	Cu(OAc) ₂ ·H ₂ O (20)	ZnI ₂ (40)	1,2-DCB	86
20	Cu(OAc)₂·H₂O (10)	ZnI₂ (40)	1,2-DCB	88
21	Cu(OAc) ₂ ·H ₂ O (10)	ZnI ₂ (40)	1,2-DCB	85 ^b , 77 ^c
22	Cu(OAc) ₂ ·H ₂ O (10)	ZnI ₂ (40)	1,2-DCB	53 ^d , 76 ^e , 0 ^f

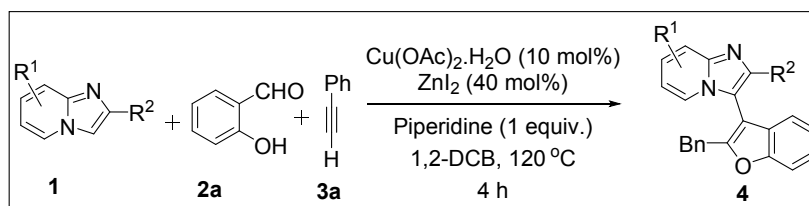
^a Reaction conditions: Carried out with 0.2 mmol of **1a**, 0.2 mmol of **2a**, piperidine (1 equiv.) and 0.21 mmol of **3a** in presence of catalyst in 2 mL of solvent for 4 h at 120 °C. ^b Temperature at 140 °C. ^c Temperature at 80 °C for 12 h with 20 mol% catalyst. ^d 1 equiv. pyrrolidine. ^e 1 equiv. morpholine. ^f 1 equiv. 1,4-tetrahydroisoquinoline.

3. Substrates scope of other heterocycles:



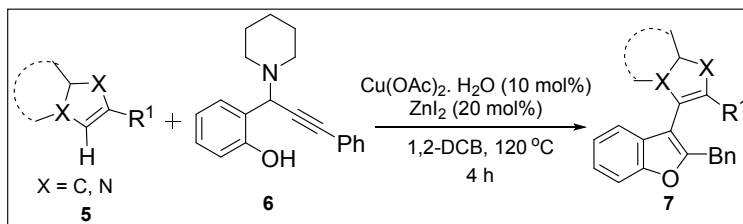
4. Experimental procedures:

4.1. Typical experimental procedure for the synthesized compounds (**4aa-4ak**):



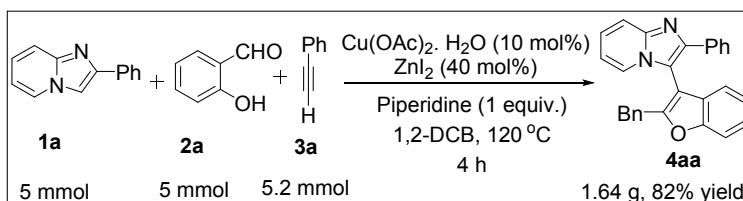
A mixture of salicylaldehyde (0.2 mmol, 24 mg) (**2a**), piperidine (1 equiv., 17 mg), phenylacetylene (0.21 mmol, 21 mg) (**3a**), and 2-phenylimidazo[1,2-*a*]pyridine (0.2 mmol, 38 mg) (**1a**) was taken in an oven dried reaction tube. Then Zinc iodide (40 mol%, 25.5 mg), and copper(II) acetate monohydrate (10 mol%, 4 mg) was added to it in 1,2-DCB (2 ml) and stirred at 120 °C for 4 h under open atmosphere. After completion of the reaction (TLC) the reaction was cooled to room temperature and extracted with dichloromethane. The organic phase was dried over anhydrous Na_2SO_4 . The crude residue was obtained after evaporating the solvent in vacuum and was purified by column chromatography on silica gel using a mixture petroleum ether and ethyl acetate (75:25) as an eluting solvent to afford the pure product (**4aa**) (70 mg, 88%) as a white solid.

4. 2. Typical experimental procedure for the synthesized compounds (4aI-7da):



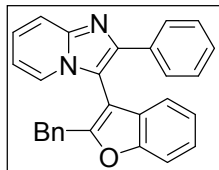
A mixture of 6-phenylimidazo[2,1-*b*]thiazole (0.2 mmol, 40 mg) (**5a**) and 2-(3-phenyl-1-(piperidin-1-yl)prop-2-yn-1-yl)phenol (0.2 mmol, 58.2 mg) (**6**) was taken in an oven dried reaction tube in presence of zinc iodide (20 mol%, 12.7 mg), and copper(II) acetate monohydrate (10 mol%, 4 mg) in 1,2-DCB (2 mL) and stirred at 120 °C for 4 h under open atmosphere. After completion of the reaction (TLC) the reaction was cooled to room temperature and extracted with dichloromethane. The organic phase was dried over anhydrous Na₂SO₄. The crude residue was obtained after evaporating the solvent in vacuum and was purified by column chromatography on silica gel using a mixture petroleum ether and ethyl acetate (76:24) as an eluting solvent to afford the pure product (**7aa**) (71%, 57 mg) as a yellow gummy mass.

4.3. Synthesis of 4aa on 5 mmol scale.

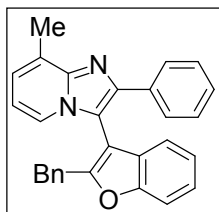


A mixture of salicylaldehyde (5 mmol, 610 mg) (**2a**), piperidine (5 equiv., 425 mg), phenylacetylene (5.2 mmol, 531 mg) (**3a**), and 2-phenylimidazo[1,2-*a*]pyridine (5 mmol, 970.4 mg) (**1a**) was taken in an oven dried 25 mL round bottom flask. Then Zinc iodide (40 mol%, 638.4 mg), and copper(II) acetate monohydrate (10 mol%, 99.8 mg) was added to it in 1,2-DCB (10 mL) and stirred at 120 °C for 4 h under open atmosphere. After completion of the reaction (TLC) the reaction was cooled to room temperature and extracted with dichloromethane. The organic phase was dried over anhydrous Na₂SO₄. The crude residue was obtained after evaporating the solvent in vacuum and was purified by column chromatography on silica gel using a mixture petroleum ether and ethyl acetate (75:25) as an eluting solvent to afford the pure product (**4aa**) (1.64 mg, 82%) as a white solid.

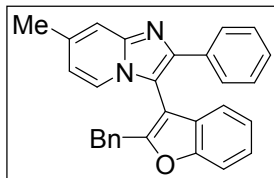
5. Characterization data for the synthesized products:



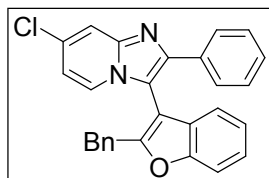
3-(2-Benzylbenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4aa): White solid (88%, 70 mg); $R_f = 0.50$ (PE : EA = 75 : 25); M.p. 130-131 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.93-7.89 (m, 3H), 7.79 (d, $J = 6.4$ Hz, 1H), 7.73 (d, $J = 8.4$ Hz, 1H), 7.53-7.48 (m, 1H), 7.44-7.39 (m, 4H), 7.37-7.35 (m, 2H), 7.26-7.23 (m, 3H), 7.07-7.05 (m, 2H), 6.85 (t, $J = 6.8$ Hz, 1H), 4.02-3.92 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.3, 154.9, 145.7, 144.4, 136.1, 133.8, 128.7, 128.5, 128.4, 128.1, 127.8, 127.7, 126.7, 125.3, 124.7, 124.1, 123.4, 119.9, 117.8, 112.6, 111.8, 111.2, 105.5, 33.6. Anal. Calcd for $\text{C}_{28}\text{H}_{20}\text{N}_2\text{O}$: C, 83.98; H, 5.03; N, 7.00%; Found: C, 84.19; H, 5.07; N, 6.89%.



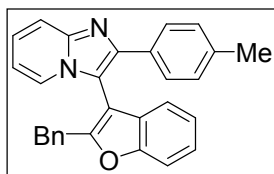
3-(2-Benzylbenzofuran-3-yl)-8-methyl-2-phenylimidazo[1,2-*a*]pyridine (4ba): Brown gummy mass (89%, 73 mg); $R_f = 0.55$ (PE : EA = 93 : 7); ^1H NMR (400 MHz, CDCl_3): δ 7.77-7.75 (m, 2H), 7.53 (t, $J = 8.4$ Hz, 2H), 7.34-7.23 (m, 4H), 7.19 (d, $J = 4.0$ Hz, 2H), 7.08-7.07 (m, 3H), 7.03 (d, $J = 7.2$ Hz, 1H), 6.90-6.88 (m, 2H), 6.60 (t, $J = 6.8$ Hz, 1H), 3.84-3.72 (m, 2H), 2.75 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.1, 155.0, 146.3, 144.1, 136.3, 134.3, 128.7, 128.6, 128.4, 128.2, 127.77, 127.71, 126.6, 124.6, 123.9, 123.4, 122.0, 120.0, 112.4, 111.7, 111.6, 106.1, 33.6, 17.2; Anal. Calcd for $\text{C}_{29}\text{H}_{22}\text{N}_2\text{O}$: C, 84.03; H, 5.35; N, 6.76%; Found: C, 83.88; H, 5.41; N, 6.68%.



3-(2-Benzylbenzofuran-3-yl)-7-methyl-2-phenylimidazo[1,2-*a*]pyridine (4ca): Brown gummy mass (87%, 72 mg); $R_f = 0.45$ (PE : EA = 76 : 24); ^1H NMR (400 MHz, CDCl_3): δ 7.71 (s, 3H), 7.53 (d, $J = 8.0$ Hz, 2H), 7.32 (t, $J = 7.6$ Hz, 1H), 7.21-7.13 (m, 5H), 7.08-7.06 (m, 3H), 6.86-6.83 (m, 2H), 6.56 (d, $J = 6.8$ Hz, 1H), 3.77-3.67 (m, 2H), 2.44 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.2, 154.9, 146.0, 143.8, 136.8, 136.1, 133.6, 128.7, 128.47, 128.40, 128.1, 127.8, 127.7, 126.6, 124.6, 123.4, 123.3, 119.8, 116.2, 115.5, 111.8, 110.7, 105.3, 33.5, 21.5; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{29}\text{H}_{23}\text{N}_2\text{O}$: 415.1805; found: 415.1795.

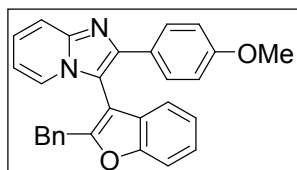


3-(2-Benzylbenzofuran-3-yl)-7-chloro-2-phenylimidazo[1,2-*a*]pyridine (4da): White solid (89%, 77mg); $R_f = 0.45$ (PE : EA = 78 : 22); M.p. 154-155 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.74-7.72 (m, 2H), 7.63 (d, $J = 9.6$ Hz, 1H), 7.57 (d, $J = 8.4$ Hz, 1H), 7.52 (s, 1H), 7.36-7.32 (m, 1H), 7.27-7.22 (m, 3H), 7.20-7.15 (m, 3H), 7.06-7.04 (m, 3H), 6.88-6.86 (m, 2H), 3.85-3.77 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.6, 155.0, 145.5, 144.1, 135.8, 133.6, 128.6, 128.4, 128.1, 128.0, 127.5, 126.8, 126.4, 124.9, 123.7, 121.9, 120.7, 119.8, 117.9, 111.88, 111.80, 105.2, 33.9. Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{ClN}_2\text{O}$: C, 77.33; H, 4.40; N, 6.44%; Found: C, 77.08; H, 4.35; N, 6.51%.

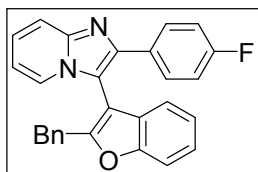


3-(2-Benzylbenzofuran-3-yl)-2-(*p*-tolyl)imidazo[1,2-*a*]pyridine (4ea): Brown gummy mass (73%, 60 mg); $R_f = 0.40$ (PE : EA = 77 : 23); ^1H NMR (400 MHz, CDCl_3): δ 7.76 (d, $J = 9.2$ Hz, 1H), 7.63-7.61 (m, 3H), 7.55 (d, $J = 8.0$ Hz, 1H), 7.36-7.31 (m, 1H), 7.26-7.17 (m, 3H), 7.08-7.04 (m, 5H), 6.89-6.87 (m, 2H), 6.69 (t, $J = 6.8$ Hz, 1H), 3.83-3.74 (m, 2H), 2.30 (s, 3H);

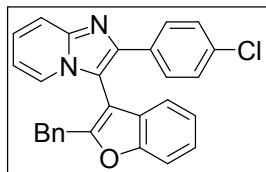
$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.3, 155.0, 145.7, 144.6, 137.6, 136.2, 131.0, 129.3, 128.7, 128.4, 128.2, 127.5, 126.6, 125.1, 124.6, 124.1, 123.4, 120.0, 117.6, 112.4, 111.8, 110.9, 105.8, 33.6, 21.3; Anal. Calcd for $\text{C}_{29}\text{H}_{22}\text{N}_2\text{O}$: C, 84.03; H, 5.35; N, 6.76%; Found: C, 84.23; H, 5.39; N, 6.65%.



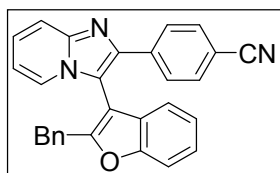
3-(2-Benzylbenzofuran-3-yl)-2-(4-methoxyphenyl)imidazo[1,2-*a*]pyridine (4fa): Brown gummy mass (71%, 61 mg); R_f = 0.50 (PE : EA = 76 : 24); ^1H NMR (400 MHz, CDCl_3): δ 7.81 (d, J = 9.2 Hz, 1H), 7.65-7.60 (m, 3H), 7.55 (d, J = 8.4 Hz, 1H), 7.35-7.31 (m, 1H), 7.27-7.23 (m, 1H), 7.21-7.16 (m, 2H), 7.09-7.06 (m, 3H), 6.89-6.87 (m, 2H), 6.75 (d, J = 8.4 Hz, 2H), 6.69 (t, J = 6.8 Hz, 1H), 3.79-3.73 (m, 5H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 159.4, 157.3, 155.0, 145.7, 144.3, 136.1, 128.9, 128.7, 128.6, 128.4, 128.1, 126.6, 126.3, 125.1, 124.6, 124.1, 123.4, 119.9, 117.5, 113.9, 112.4, 111.8, 105.7, 55.2, 33.6; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{29}\text{H}_{23}\text{N}_2\text{O}_2$: 431.1760; found: 431.1755.



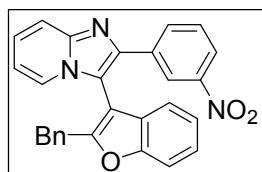
3-(2-Benzylbenzofuran-3-yl)-2-(4-fluorophenyl)imidazo[1,2-*a*]pyridine (4ga): Brown gummy mass (84%, 70 mg); R_f = 0.55 (PE : EA = 74 : 26); ^1H NMR (400 MHz, CDCl_3): δ 7.73-7.69 (m, 3H), 7.61 (d, J = 7.2 Hz, 1H), 7.56 (d, J = 8.4 Hz, 1H), 7.36-7.31 (m, 1H), 7.25-7.15 (m, 3H), 7.09-7.06 (m, 3H), 6.96-6.89 (m, 4H), 6.68 (t, J = 6.8 Hz, 1H), 3.86-3.76 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 162.6 ($J_{\text{C-F}}$ = 245 Hz), 157.3, 155.0, 145.8, 143.7, 136.0, 130.2 ($J_{\text{C-F}}$ = 3 Hz), 129.2 ($J_{\text{C-F}}$ = 8 Hz), 128.6, 128.4, 128.0, 126.7, 125.2, 124.8, 124.1, 123.5, 119.9, 117.6, 115.5 ($J_{\text{C-F}}$ = 21 Hz), 112.4, 111.8, 110.9, 105.6, 33.6; Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{FN}_2\text{O}$: C, 80.37; H, 4.58; N, 6.69%; Found: C, 80.21; H, 4.51; N, 6.60%.



3-(2-Benzylbenzofuran-3-yl)-2-(4-chlorophenyl)imidazo[1,2-*a*]pyridine (4ha): Light yellow gummy mass (78%, 67 mg); $R_f = 0.45$ (PE : EA = 80 : 20); ^1H NMR (400 MHz, CDCl_3): δ 7.66 (d, $J = 8.8$ Hz, 1H), 7.59-7.56 (m, 2H), 7.53-7.48 (m, 2H), 7.28-7.23 (m, 1H), 7.19-7.06 (m, 5H), 7.00-6.96 (m, 3H), 6.82-6.79 (m, 2H), 6.62-6.59 (m, 1H), 3.78-3.67 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.2, 154.9, 145.8, 143.3, 135.9, 133.7, 132.5, 128.79, 128.76, 128.6, 128.4, 127.9, 126.7, 125.4, 124.8, 124.1, 123.5, 119.8, 117.6, 112.6, 111.8, 111.3, 105.4, 33.6; Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{ClN}_2\text{O}$: C, 77.33; H, 4.40; N, 6.44%; Found: C, 77.55; H, 4.46; N, 6.32%.

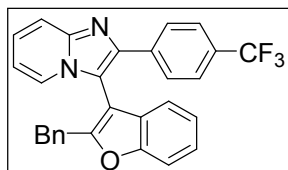


4-(3-(2-Benzylbenzofuran-3-yl)imidazo[1,2-*a*]pyridin-2-yl)benzonitrile (4ia): White solid (81%, 68 mg); $R_f = 0.45$ (PE : EA = 74 : 26); M.p. 145-146 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.82-7.80 (m, 2H), 7.72-7.69 (m, 1H), 7.62-7.58 (m, 2H), 7.48-7.46 (m, 2H), 7.38-7.34 (m, 1H), 7.29-7.18 (m, 2H), 7.13 (d, $J = 6.8$ Hz, 1H), 7.10-7.02 (m, 3H), 6.88-6.86 (m, 2H), 6.74-6.70 (m, 1H), 3.88-3.76 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.3, 155.0, 146.0, 142.3, 138.6, 135.6, 132.3, 128.5, 127.7, 126.8, 125.9, 125.0, 124.3, 123.7, 119.7, 119.1, 117.9, 112.9, 112.6, 111.9, 111.0, 105.2, 33.7; Anal. Calcd for $\text{C}_{29}\text{H}_{19}\text{N}_3\text{O}$: C, 81.86; H, 4.50; N, 9.88%; Found: C, 82.04; H, 4.47; N, 9.98%.



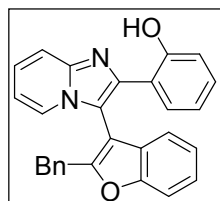
3-(2-Benzylbenzofuran-3-yl)-2-(3-nitrophenyl)imidazo[1,2-*a*]pyridine (4ja): Light yellow gummy mass (79%, 70 mg); $R_f = 0.50$ (PE : EA = 74 : 26); ^1H NMR (400 MHz, CDCl_3): δ 8.66 (t, $J = 2.0$ Hz, 1H), 8.04-8.01 (m, 1H), 7.97-7.95 (m, 1H), 7.75-7.72 (m, 1H), 7.66-7.60 (m, 2H), 7.39-7.27 (m, 3H), 7.23-7.19 (m, 1H), 7.14 (d, $J = 6.8$ Hz, 1H), 7.06-7.02 (m, 3H), 6.92-6.89 (m,

2H), 6.76-6.72 (m, 1H), 3.94-3.82 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.4, 155.0, 148.6, 145.9, 142.0, 135.9, 135.6, 132.9, 129.3, 128.4, 127.7, 126.8, 125.8, 125.0, 124.3, 123.7, 122.3, 119.7, 117.9, 112.9, 112.2, 111.9, 105.2, 33.8; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{28}\text{H}_{20}\text{N}_3\text{O}_3$: 446.1505; found: 446.1500.



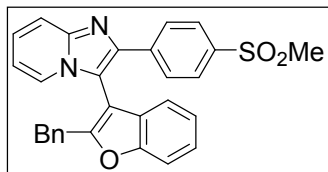
3-(2-Benzylbenzofuran-3-yl)-2-(4-(trifluoromethyl)phenyl)imidazo[1,2-a]pyridine (4ka):

Brown gummy mass (78%, 73 mg); R_f = 0.50 (PE : EA = 76 : 24); ^1H NMR (400 MHz, CDCl_3): δ 7.84 (d, J = 8.0 Hz, 2H), 7.73 (d, J = 9.2 Hz, 1H), 7.64-7.58 (m, 2H), 7.49 (d, J = 8.4 Hz, 2H), 7.38-7.34 (m, 1H), 7.30-7.24 (m, 1H), 7.22-7.16 (m, 2H), 7.07-7.03 (m, 3H), 6.88-6.86 (m, 2H), 6.74-6.70 (m, 1H), 3.88-3.77 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.4, 155.0, 146.0, 143.0, 137.6, 135.8, 129.7, 129.4, 128.6, 128.5, 128.0, 127.6, 126.8, 125.5 ($J_{\text{C-F}}$ = 12 Hz, 9 Hz), 124.9, 124.3, 123.7, 119.8, 117.9, 112.8, 112.2, 111.9, 105.4, 33.7; Anal. Calcd for $\text{C}_{29}\text{H}_{19}\text{F}_3\text{N}_2\text{O}$: C, 74.35; H, 4.09; N, 5.98%; Found: C, 74.58; H, 4.03; N, 6.09%.



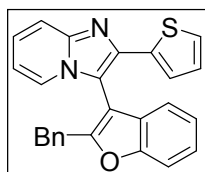
2-(3-(2-Benzylbenzofuran-3-yl)imidazo[1,2-a]pyridin-2-yl)phenol (4la):

White solid (85%, 70 mg); R_f = 0.45 (PE : EA = 81 : 19); M.p. 210-211 $^\circ\text{C}$; ^1H NMR (400 MHz, CDCl_3): δ 13.15 (br s, 1H), 7.67-7.64 (m, 1H), 7.61-7.56 (m, 2H), 7.38-7.34 (m, 1H), 7.31-7.27 (m, 2H), 7.22-7.14 (m, 3H), 7.07-7.05 (m, 4H), 6.95-6.93 (m, 2H), 6.74-6.70 (m, 1H), 6.57-6.53 (m, 1H), 3.97-3.87 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.0, 157.8, 155.1, 143.38, 143.32, 135.8, 129.6, 128.7, 128.4, 127.9, 126.79, 126.70, 125.7, 124.9, 123.8, 123.6, 119.9, 118.9, 117.7, 116.7, 113.0, 111.8, 109.9, 105.7, 34.0; Anal. Calcd for $\text{C}_{28}\text{H}_{20}\text{N}_2\text{O}_2$: C, 80.75; H, 4.84; N, 6.73%; Found: C, 80.60; H, 4.89; N, 6.65%.



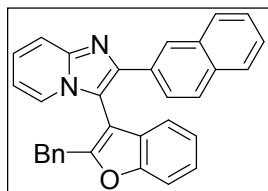
3-(2-Benzylbenzofuran-3-yl)-2-(4-(methylsulfonyl)phenyl)imidazo[1,2-a]pyridine (4ma):

Yellow gummy mass (70%, 66 mg); R_f = 0.50 (PE : EA = 55 : 45); ^1H NMR (400 MHz, CDCl_3): δ 7.92 (d, J = 8.8 Hz, 2H), 7.79-7.75 (m, 3H), 7.61 (t, J = 8.4 Hz, 2H), 7.39-7.35 (m, 1H), 7.33-7.28 (m, 1H), 7.22 (t, J = 7.6 Hz, 1H), 7.14 (d, J = 8.0 Hz, 1H), 7.08-7.02 (m, 3H), 6.89-6.87 (m, 2H), 6.76-6.72 (m, 1H), 3.88-3.78 (m, 2H), 3.01 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.5, 155.0, 146.0, 142.2, 139.5, 139.2, 135.6, 130.0, 128.5, 128.1, 127.8, 127.6, 127.2, 126.9, 126.1, 125.1, 124.3, 123.8, 119.7, 117.9, 113.1, 112.0, 105.1, 44.6, 33.8; Anal. Calcd for $\text{C}_{29}\text{H}_{22}\text{N}_2\text{O}_3\text{S}$: C, 72.78; H, 4.63; N, 5.85%; Found: C, 72.99; H, 4.59; N, 5.78%.



3-(2-Benzylbenzofuran-3-yl)-2-(thiophen-2-yl)imidazo[1,2-a]pyridine (4na):

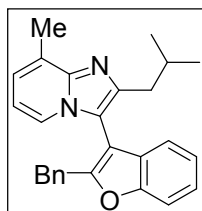
White solid (86%, 69 mg); R_f = 0.50 (PE : EA = 80 : 20); M.p. 179-180 $^\circ\text{C}$; ^1H NMR (400 MHz, CDCl_3): δ 7.70 (d, J = 8.8 Hz, 1H), 7.58-7.54 (m, 2H), 7.35-7.30 (m, 1H), 7.24-7.18 (m, 4H), 7.16-7.13 (m, 1H), 7.10-7.08 (m, 3H), 7.00-6.98 (m, 2H), 6.93-6.91 (m, 1H), 6.65 (t, J = 6.8 Hz, 1H), 3.95 (s, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.9, 155.0, 145.6, 140.0, 137.1, 136.0, 128.6, 128.4, 127.9, 127.6, 126.7, 125.6, 125.2, 124.8, 124.6, 123.9, 123.4, 119.9, 117.3, 112.4, 111.6, 110.0, 105.0, 33.9; Anal. Calcd for $\text{C}_{26}\text{H}_{18}\text{N}_2\text{OS}$: C, 76.82; H, 4.46; N, 6.89%; Found: C, 76.63; H, 4.39; N, 6.99%.



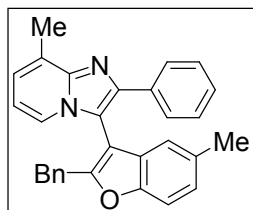
3-(2-Benzylbenzofuran-3-yl)-2-(naphthalen-2-yl)imidazo[1,2-a]pyridine (4oa):

Brown gummy mass (78%, 70 mg); R_f = 0.45 (PE : EA = 78 : 22); ^1H NMR (400 MHz, CDCl_3): δ 8.38 (s, 1H), 7.79-7.74 (m, 4H), 7.69-7.65 (m, 2H), 7.59 (d, J = 8.4 Hz, 1H), 7.46-7.40 (m, 2H), 7.38-

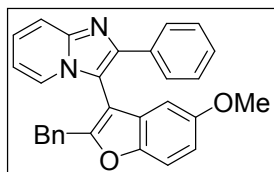
7.34 (m, 1H), 7.29-7.20 (m, 3H), 7.03-6.95 (m, 3H), 6.87-6.85 (m, 2H), 6.70-6.69 (m, 1H), 3.86-3.75 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.4, 155.0, 145.9, 144.4, 136.0, 133.6, 133.0, 131.5, 128.6, 128.5, 128.4, 128.3, 128.1, 127.6, 126.8, 126.6, 126.1, 125.35, 125.32, 124.8, 124.7, 124.2, 123.5, 120.0, 117.6, 112.4, 111.8, 111.6, 105.9, 33.7; Anal. Calcd for $\text{C}_{32}\text{H}_{22}\text{N}_2\text{O}$: C, 85.31; H, 4.92; N, 6.22%; Found: C, 85.11; H, 4.89; N, 6.30%.



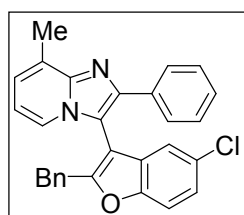
3-(2-Benzylbenzofuran-3-yl)-2-isobutyl-8-methylimidazo[1,2-a]pyridine (4pa): Brown gummy mass (90%, 70 mg); R_f = 0.60 (PE : EA = 81 : 19); ^1H NMR (400 MHz, CDCl_3): δ 7.53 (d, J = 8.4 Hz, 1H), 7.47 (d, J = 6.8 Hz, 1H), 7.31-7.29 (m, 1H), 7.23-7.17 (m, 4H), 7.12 (d, J = 8.4 Hz, 3H), 7.00 (d, J = 7.2 Hz, 1H), 6.59 (t, J = 7.2 Hz, 1H), 4.06-3.97 (m, 2H), 2.74 (s, 3H), 2.72-2.67 (m, 1H), 2.53-2.48 (m, 1H), 2.25-2.14 (m, 1H), 0.86 (d, J = 6.8 Hz, 3H), 0.73 (d, J = 6.8 Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 156.9, 154.8, 145.9, 145.8, 136.6, 128.7, 128.6, 128.5, 126.84, 126.82, 124.4, 123.8, 123.2, 122.0, 119.8, 112.7, 112.0, 111.6, 105.7, 37.4, 33.5, 28.9, 22.8, 22.2, 17.7; Anal. Calcd for $\text{C}_{27}\text{H}_{26}\text{N}_2\text{O}$: C, 82.20; H, 6.64; N, 7.10%; Found: C, 82.44; H, 6.69; N, 7.19%.



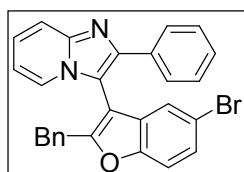
3-(2-Benzyl-5-methylbenzofuran-3-yl)-8-methyl-2-phenylimidazo[1,2-a]pyridine (4bb): Brown gummy mass (77%, 65 mg); R_f = 0.45 (PE : EA = 94 : 6); ^1H NMR (400 MHz, CDCl_3): δ 7.78-7.76 (m, 2H), 7.52 (d, J = 6.8 Hz, 1H), 7.42 (d, J = 8.4 Hz, 1H), 7.30-7.24 (m, 3H), 7.15-7.12 (m, 1H), 7.09-7.06 (m, 3H), 7.04-7.02 (m, 1H), 6.99 (s, 1H), 6.89-6.87 (m, 2H), 6.62 (t, J = 6.8 Hz, 1H), 3.81-3.70 (m, 2H), 2.75 (s, 3H), 2.35 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.2, 153.4, 146.3, 144.1, 136.4, 134.4, 133.0, 131.0, 130.6, 128.7, 128.6, 128.4, 127.7, 127.6, 126.6, 125.8, 123.7, 122.1, 119.8, 112.3, 111.8, 111.2, 105.9, 33.5, 21.4, 17.2; Anal. Calcd for $\text{C}_{30}\text{H}_{24}\text{N}_2\text{O}$: C, 84.08; H, 5.65; N, 6.54%; Found: C, 84.26; H, 5.59; N, 6.42%.



3-(2-Benzyl-5-methoxybenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ac): Brown solid (89%, 76 mg); $R_f = 0.45$ (PE : EA = 80 : 20); M.p. 135-136 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.77-7.75 (m, 2H), 7.72 (d, $J = 9.2$ Hz, 1H), 7.61 (d, $J = 6.8$ Hz, 1H), 7.43 (d, $J = 8.8$ Hz, 1H), 7.30-7.24 (m, 3H), 7.23-7.21 (m, 1H), 7.05 (t, $J = 3.6$ Hz, 3H), 6.93-6.90 (m, 1H), 6.88-6.86 (m, 2H), 6.69-6.66 (m, 1H), 6.59 (d, $J = 2.4$ Hz, 1H), 3.82-3.73 (m, 2H), 3.66 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.0, 156.5, 149.8, 145.8, 144.5, 136.2, 134.1, 128.8, 128.7, 128.6, 128.4, 127.8, 127.5, 126.6, 125.1, 124.1, 117.6, 113.4, 112.4, 112.3, 111.2, 105.9, 102.0, 55.9, 33.8; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{29}\text{H}_{23}\text{N}_2\text{O}_2$: 431.1760; found: 431.1756.

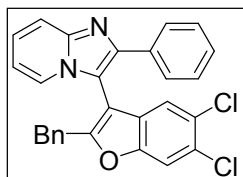


3-(2-Benzyl-5-chlorobenzofuran-3-yl)-8-methyl-2-phenylimidazo[1,2-*a*]pyridine (4bd): Light yellow gummy mass (83%, 74 mg); $R_f = 0.50$ (PE : EA = 92 : 8); ^1H NMR (400 MHz, CDCl_3): δ 7.74-7.72 (m, 2H), 7.48-7.43 (m, 2H), 7.30-7.24 (m, 4H), 7.16 (d, $J = 2.0$ Hz, 1H), 7.09-7.03 (m, 4H), 6.88-6.85 (m, 2H), 6.63 (t, $J = 6.8$ Hz, 1H), 3.82-3.71 (m, 2H), 2.74 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.8, 153.4, 146.4, 144.5, 135.9, 134.3, 129.8, 129.2, 128.8, 128.6, 128.5, 127.9, 127.8, 127.7, 126.8, 124.9, 123.9, 121.8, 119.5, 112.8, 112.6, 110.6, 106.0, 33.6, 17.2; Anal. Calcd for $\text{C}_{29}\text{H}_{21}\text{ClN}_2\text{O}$: C, 77.59; H, 4.72; N, 6.24%; Found: C, 77.38; H, 4.76; N, 6.11%.

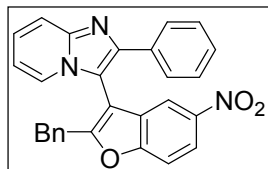


3-(2-Benzyl-5-bromobenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ae): Brown solid (73%, 69 mg); $R_f = 0.50$ (PE : EA = 82 : 18); M.p. 223-224 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.72-7.58 (m, 4H), 7.43-7.41 (m, 2H), 7.31-7.27 (m, 5H), 7.07-7.04 (m, 3H), 6.86-6.84 (m, 2H),

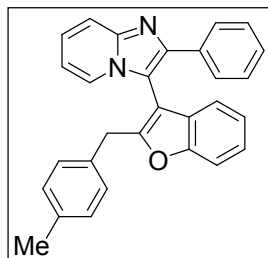
6.72 (t, $J = 6.8$ Hz, 1H), 3.82-3.73 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.9, 153.7, 145.9, 144.8, 135.7, 133.8, 130.2, 128.7, 128.6, 128.5, 128.0, 127.7, 127.5, 126.8, 125.3, 123.9, 122.4, 117.8, 116.7, 113.3, 112.7, 110.2, 105.4, 33.6; Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{BrN}_2\text{O}$: C, 70.16; H, 4.00; N, 5.84%; Found: C, 70.33; H, 3.97; N, 5.77%.



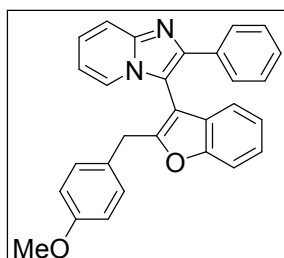
3-(2-Benzyl-5,6-dichlorobenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4af): White solid (75%, 70 mg); $R_f = 0.45$ (PE : EA = 77 : 23); M.p. 165-166 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.71-7.68 (m, 3H), 7.48 (d, $J = 6.8$ Hz, 1H), 7.33 (d, $J = 2.0$ Hz, 1H), 7.30-7.22 (m, 4H), 7.04-7.00 (m, 4H), 6.86-6.84 (m, 2H), 6.68 (t, $J = 6.8$ Hz, 1H), 3.83 (s, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 160.0, 149.4, 145.9, 145.0, 135.2, 133.7, 130.8, 129.6, 128.7, 128.6, 128.5, 128.1, 127.4, 126.9, 125.4, 125.0, 123.7, 118.1, 117.8, 117.7, 112.7, 109.5, 106.4, 33.7; Anal. Calcd for $\text{C}_{28}\text{H}_{18}\text{Cl}_2\text{N}_2\text{O}$: C, 71.65; H, 3.87; N, 5.97%; Found: C, 71.84; H, 3.82; N, 6.05%.



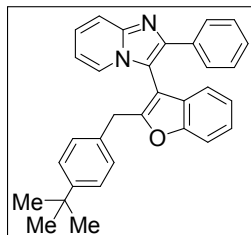
3-(2-Benzyl-5-nitrobenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ag): Light yellow solid (92%, 81 mg); $R_f = 0.55$ (PE : EA = 76 : 24); M.p. 184-185 °C; ^1H NMR (400 MHz, CDCl_3): δ 8.28-8.25 (m, 1H), 8.10 (d, $J = 2.4$ Hz, 1H), 7.76 (d, $J = 8.8$ Hz, 1H), 7.70-7.68 (m, 2H), 7.64 (d, $J = 9.2$ Hz, 1H), 7.57 (d, $J = 6.8$ Hz, 1H), 7.31-7.27 (m, 4H), 7.10-7.06 (m, 3H), 6.90-6.88 (m, 2H), 6.74 (t, $J = 6.8$ Hz, 1H), 3.90-3.81 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 161.0, 157.7, 146.1, 145.3, 144.7, 135.1, 133.7, 128.9, 128.7, 128.6, 128.2, 127.5, 127.0, 125.5, 123.6, 120.7, 117.9, 116.2, 113.0, 112.3, 109.0, 106.9, 33.8; Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{N}_3\text{O}_3$: C, 75.49; H, 4.30; N, 9.43%; Found: C, 75.29; H, 4.36; N, 9.54%.



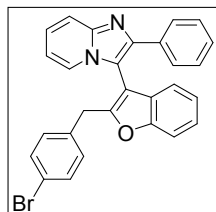
3-(2-(4-Methylbenzyl)benzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ah): Brown gummy mass (79%, 65 mg); R_f = 0.50 (PE : EA = 78 : 22); ^1H NMR (400 MHz, CDCl_3): δ 7.75 (d, J = 8.0 Hz, 3H), 7.62 (d, J = 6.8 Hz, 1H), 7.55 (d, J = 8.0 Hz, 1H), 7.35-7.31 (m, 1H), 7.26-7.22 (m, 4H), 7.20-7.16 (m, 2H), 6.87 (d, J = 7.6 Hz, 2H), 6.77 (d, J = 7.6 Hz, 2H), 6.68 (t, J = 6.8 Hz, 1H), 3.80-3.71 (m, 2H), 2.21 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.6, 155.0, 145.8, 144.6, 136.3, 134.1, 133.1, 129.3, 129.1, 128.6, 128.2, 127.8, 127.6, 125.1, 124.6, 124.2, 123.4, 119.9, 117.7, 112.4, 111.8, 111.3, 105.6, 33.3, 21.0; Anal. Calcd for $\text{C}_{29}\text{H}_{22}\text{N}_2\text{O}$: C, 84.03; H, 5.35; N, 6.76%; Found: C, 84.26; H, 5.40; N, 6.67%.



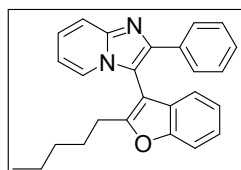
3-(2-(4-Methoxybenzyl)benzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ai): Light yellow gummy mass (66%, 56 mg); R_f = 0.45 (PE : EA = 89 : 19); ^1H NMR (400 MHz, CDCl_3): δ 7.78-7.72 (m, 3H), 7.61 (d, J = 9.2 Hz, 1H), 7.55 (d, J = 8.4 Hz, 1H), 7.35-7.31 (m, 1H), 7.27-7.22 (m, 4H), 7.20-7.16 (m, 2H), 6.80-6.77 (m, 2H), 6.71-6.68 (m, 1H), 6.61-6.58 (m, 2H), 3.73 (d, J = 5.6 Hz, 2H), 3.69 (s, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.3, 157.7, 155.0, 145.8, 144.5, 134.0, 129.7, 128.6, 128.27, 128.21, 128.1, 127.8, 127.6, 125.1, 124.6, 124.2, 123.4, 119.9, 117.7, 113.8, 112.4, 111.8, 105.4, 55.3, 32.8; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{29}\text{H}_{23}\text{N}_2\text{O}_2$: 431.1760; found: 431.1758.



3-(2-(4-(*tert*-Butyl)benzyl)benzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4aj): Brown gummy mass (65%, 59 mg); $R_f = 0.45$ (PE : EA = 77 : 23); ^1H NMR (400 MHz, CDCl_3): δ 7.75 (br s, 3H), 7.62-7.56 (m, 2H), 7.35-7.31 (m, 1H), 7.27-7.22 (m, 4H), 7.19 (d, $J = 6.4$ Hz, 2H), 7.07 (d, $J = 8.4$ Hz, 2H), 6.81 (d, $J = 8.4$ Hz, 2H), 6.67 (t, $J = 6.8$ Hz, 1H), 3.78 (s, 2H), 1.21 (s, 9H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.6, 155.0, 149.5, 134.2, 133.1, 128.6, 128.3, 127.8, 127.5, 125.3, 125.0, 124.6, 124.2, 123.4, 119.9, 117.7, 113.7, 112.3, 111.7, 105.7, 34.4, 33.2, 31.4; Anal. Calcd for $\text{C}_{32}\text{H}_{28}\text{N}_2\text{O}$: C, 84.18; H, 6.18; N, 6.14%; Found: C, 84.00; H, 6.12; N, 6.24%.

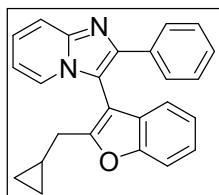


3-(2-(4-Bromobenzyl)benzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4ak): Brown gummy mass (67%, 64mg); $R_f = 0.40$ (PE : EA = 80 : 20); ^1H NMR (400 MHz, CDCl_3): δ 7.85-7.83 (m, 3H), 7.72-7.66 (m, 2H), 7.48-7.44 (m, 1H), 7.37-7.32 (m, 6H), 7.28-7.25 (m, 2H), 6.84-6.79 (m, 3H), 3.89-3.79 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 156.5, 155.0, 147.9, 144.8, 135.1, 134.1, 131.5, 130.4, 128.7, 128.1, 127.9, 127.6, 125.2, 124.9, 124.1, 123.6, 120.6, 120.0, 117.7, 112.5, 111.8, 106.1, 33.1; Anal. Calcd for $\text{C}_{28}\text{H}_{19}\text{BrN}_2\text{O}$: C, 70.16; H, 4.00; N, 5.84%; Found: C, 70.37; H, 4.03; N, 5.77%.

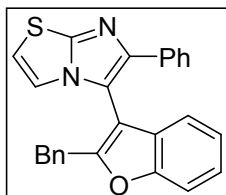


3-(2-Pentylbenzofuran-3-yl)-2-phenylimidazo[1,2-*a*]pyridine (4al): Yellow gummy mass (86%, 65mg); $R_f = 0.45$ (PE : EA = 88 : 12); ^1H NMR (400 MHz, CDCl_3): δ 7.80 (d, $J = 8.8$ Hz, 1H), 7.72-7.70 (m, 3H), 7.57 (d, $J = 8.0$ Hz, 1H), 7.35-7.31 (m, 1H), 7.28-7.20 (m, 4H), 7.18-

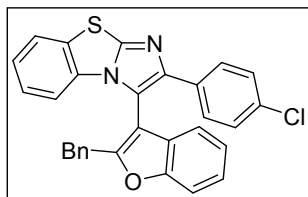
7.14 (m, 2H), 6.76-6.72 (m, 1H), 2.48-2.34 (m, 2H), 1.54-1.45 (m, 1H), 1.40-1.32 (m, 1H), 1.07-1.03 (m, 4H), 0.69 (t, $J = 7.2$ Hz, 3H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 159.5, 154.8, 145.7, 144.2, 134.0, 128.4, 128.1, 127.8, 127.6, 125.1, 124.3, 124.2, 123.3, 119.8, 117.7, 112.5, 111.8, 111.5, 104.8, 31.3, 27.1, 26.9, 22.2, 13.9; Anal. Calcd for $\text{C}_{26}\text{H}_{24}\text{N}_2\text{O}$: C, 82.07; H, 6.36; N, 7.36%; Found: C, 82.33; H, 6.41; N, 7.45%.



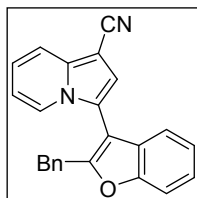
3-(2-(Cyclopropylmethyl)benzofuran-3-yl)-2-phenylimidazo[1,2-a]pyridine (4am): Brown gummy mass (83%, 60 mg); $R_f = 0.50$ (PE : EA = 87 : 13); ^1H NMR (400 MHz, CDCl_3): δ 7.77-7.71 (m, 4H), 7.60 (d, $J = 8.4$ Hz, 1H), 7.37-7.33 (m, 1H), 7.29-7.22 (m, 4H), 7.20-7.16 (m, 2H), 6.75-6.71 (m, 1H), 2.39 (d, $J = 6.8$ Hz, 2H), 0.87-0.79 (m, 1H), 0.29-0.25 (m, 2H), (-)0.08-(-)0.11 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 159.1, 154.9, 145.8, 144.3, 134.2, 128.5, 128.2, 127.8, 127.5, 125.0, 124.4, 124.2, 123.3, 119.9, 117.7, 112.4, 111.63, 111.60, 105.0, 32.1, 9.1, 4.6, 4.5; Anal. Calcd for $\text{C}_{25}\text{H}_{22}\text{N}_2\text{O}$: C, 82.39; H, 5.53; N, 7.69%; Found: C, 82.55; H, 5.60; N, 7.61%.



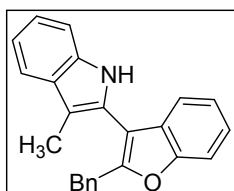
5-(2-Benzylbenzofuran-3-yl)-6-phenylimidazo[2,1-b]thiazole (7aa): yellow gummy mass (71%, 57 mg); $R_f = 0.45$ (PE : EA = 76 : 24); ^1H NMR (400 MHz, CDCl_3): δ 7.67-7.65 (m, 2H), 7.52 (d, $J = 8.0$ Hz, 1H), 7.34-7.29 (m, 1H), 7.27-7.18 (m, 5H), 7.14-7.10 (m, 3H), 6.98-6.95 (m, 3H), 6.73 (d, $J = 4.8$ Hz, 1H), 3.89-3.80 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 156.4, 154.8, 149.9, 145.7, 136.4, 134.4, 129.2, 128.8, 128.6, 128.5, 128.1, 127.3, 126.8, 124.9, 124.6, 123.4, 119.9, 117.9, 112.5, 111.7, 106.6, 33.7; Anal. Calcd for $\text{C}_{26}\text{H}_{18}\text{N}_2\text{OS}$: C, 76.82; H, 4.46; N, 6.89%; Found: C, 76.60; H, 4.42; N, 7.00%.



3-(2-Benzylbenzofuran-3-yl)-2-(4-chlorophenyl)benzo[d]imidazo[2,1-b]thiazole (7ba): Light yellow solid (62%, 60 mg); $R_f = 0.45$ (PE : EA = 92 : 7); M.p. 215-216 °C; ^1H NMR (400 MHz, CDCl_3): δ 7.93 (d, $J = 8.0$ Hz, 1H), 7.75-7.72 (m, 4H), 7.68 (t, $J = 1.2$ Hz, 1H), 7.52-7.48 (m, 2H), 7.45-7.43 (m, 2H), 7.35-7.30 (m, 1H), 7.25-7.18 (m, 2H), 7.06 (d, $J = 7.2$ Hz, 2H), 6.96 (d, $J = 8.0$ Hz, 1H), 4.11 (s, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 158.8, 153.4, 149.0, 145.5, 135.2, 133.4, 132.7, 132.4, 130.9, 130.4, 128.8, 128.7, 128.3, 128.1, 127.9, 126.8, 126.3, 124.9, 124.3, 122.6, 117.1, 113.4, 113.1, 112.8, 105.9, 33.9; Anal. Calcd for $\text{C}_{30}\text{H}_{19}\text{ClN}_2\text{OS}$: C, 73.39; H, 3.90; N, 5.71%; Found: C, 73.59; H, 3.96; N, 5.59%.

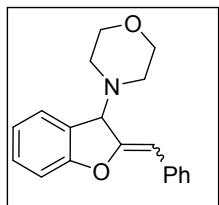


3-(2-benzylbenzofuran-3-yl)indolizine-1-carbonitrile (7ca): Yellow gummy mass (53%, 36 mg); $R_f = 0.60$ (PE : EA = 93 : 7); ^1H NMR (400 MHz, CDCl_3): δ 7.79-7.73 (m, 2H), 7.53 (d, $J = 8.4$ Hz, 1H), 7.35-7.30 (m, 1H), 7.28-7.18 (m, 4H), 7.18-7.11 (m, 4H), 7.06 (s, 1H), 6.72-6.68 (m, 1H), 4.12-4.06 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 157.0, 154.5, 138.7, 136.7, 128.8, 128.6, 127.7, 127.0, 126.1, 124.9, 124.7, 123.4, 122.7, 119.9, 118.2, 118.0, 116.9, 113.0, 111.8, 106.7, 82.1, 33.4; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{24}\text{H}_{17}\text{N}_2\text{O}$: 349.1341; found: 349.1337.

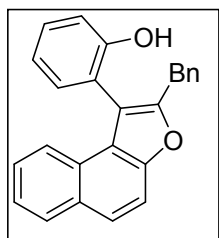


2-(2-Benzylbenzofuran-3-yl)-3-methyl-1H-indole (7da): Brown gummy mass (45%, 30 mg); $R_f = 0.45$ (PE : EA = 94 : 6); ^1H NMR (400 MHz, CDCl_3): δ 7.85 (s, 1H), 7.63 (d, $J = 7.6$ Hz, 1H), 7.48-7.44 (m, 2H), 7.34 (d, $J = 8.0$ Hz, 1H), 7.30-7.14 (m, 9H), 4.14 (s, 2H), 2.28 (s, 3H);

$^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 155.1, 154.5, 137.6, 136.4, 129.5, 129.4, 128.9, 128.8, 128.6, 126.8, 124.3, 123.1, 122.3, 120.2, 119.5, 118.9, 111.4, 111.0, 110.8, 33.6, 9.8; HRMS (ESI-TOF) m/z : $[\text{M} + \text{H}]^+$ Calcd for $\text{C}_{24}\text{H}_{20}\text{NO}$: 338.1539; found: 338.1538.



4-(2-Benzylidene-2,3-dihydrobenzofuran-3-yl)morpholine (9):² Yellow solid (73%, 42 mg); R_f = 0.45 (PE : EA = 86 : 14); M.p. 102-103 $^\circ\text{C}$; ^1H NMR (400 MHz, CDCl_3): δ 7.55-7.53 (m, 2H), 7.50-7.44 (m, 2H), 7.35-7.32 (m, 2H), 7.29-7.16 (m, 3H), 6.66 (s, 1H), 4.51 (s, 1H), 3.73 (t, J = 4.8 Hz, 4H), 2.51-2.44 (m, 4H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 156.9, 155.1, 138.7, 128.7, 128.6, 128.2, 127.9, 124.0, 122.8, 120.8, 111.5, 105.3, 69.8, 67.1, 52.3; Anal. Calcd for $\text{C}_{19}\text{H}_{19}\text{NO}_2$: C, 77.79; H, 6.53; N, 4.77%; Found: C, 78.01; H, 6.59; N, 4.68%.

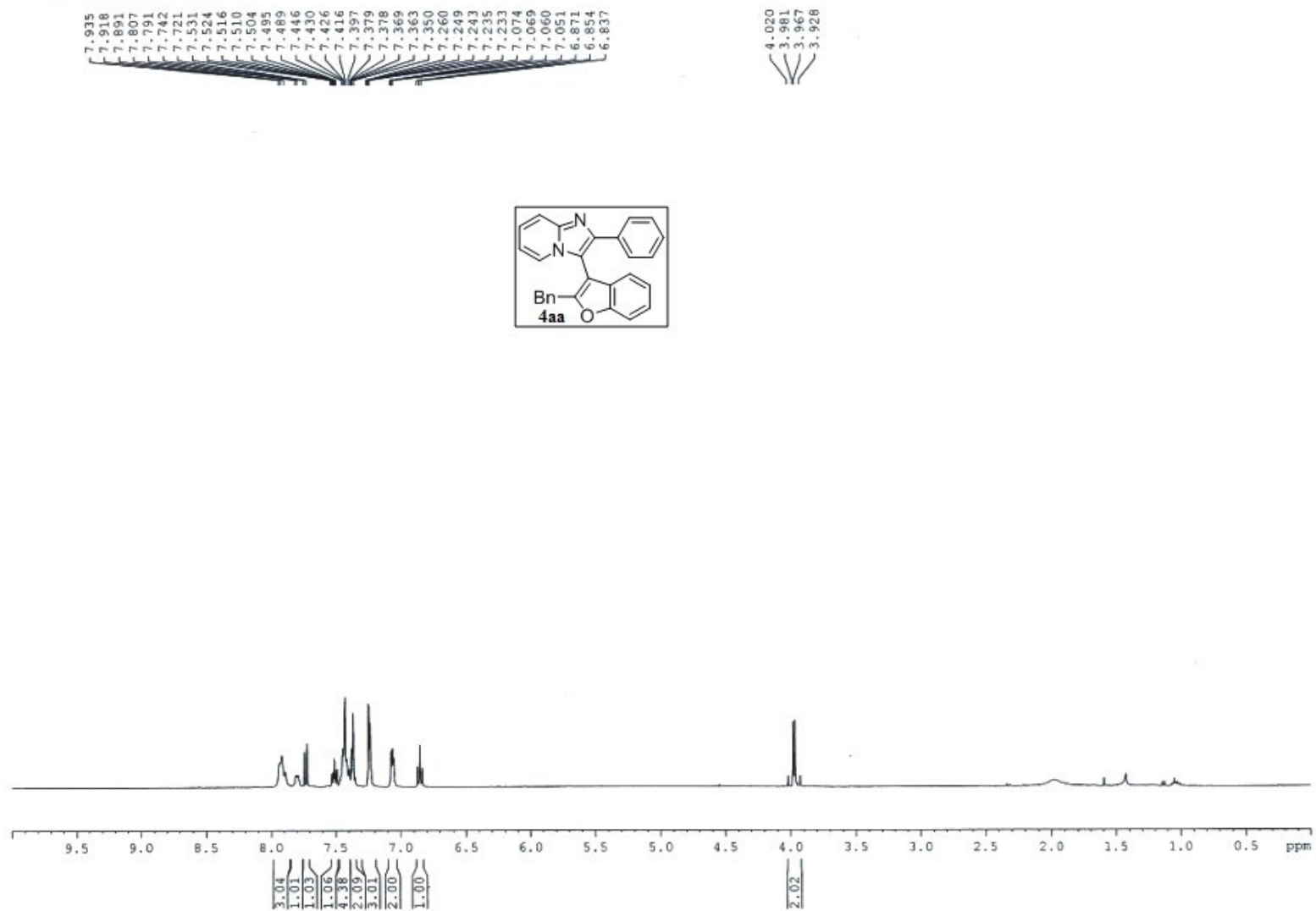


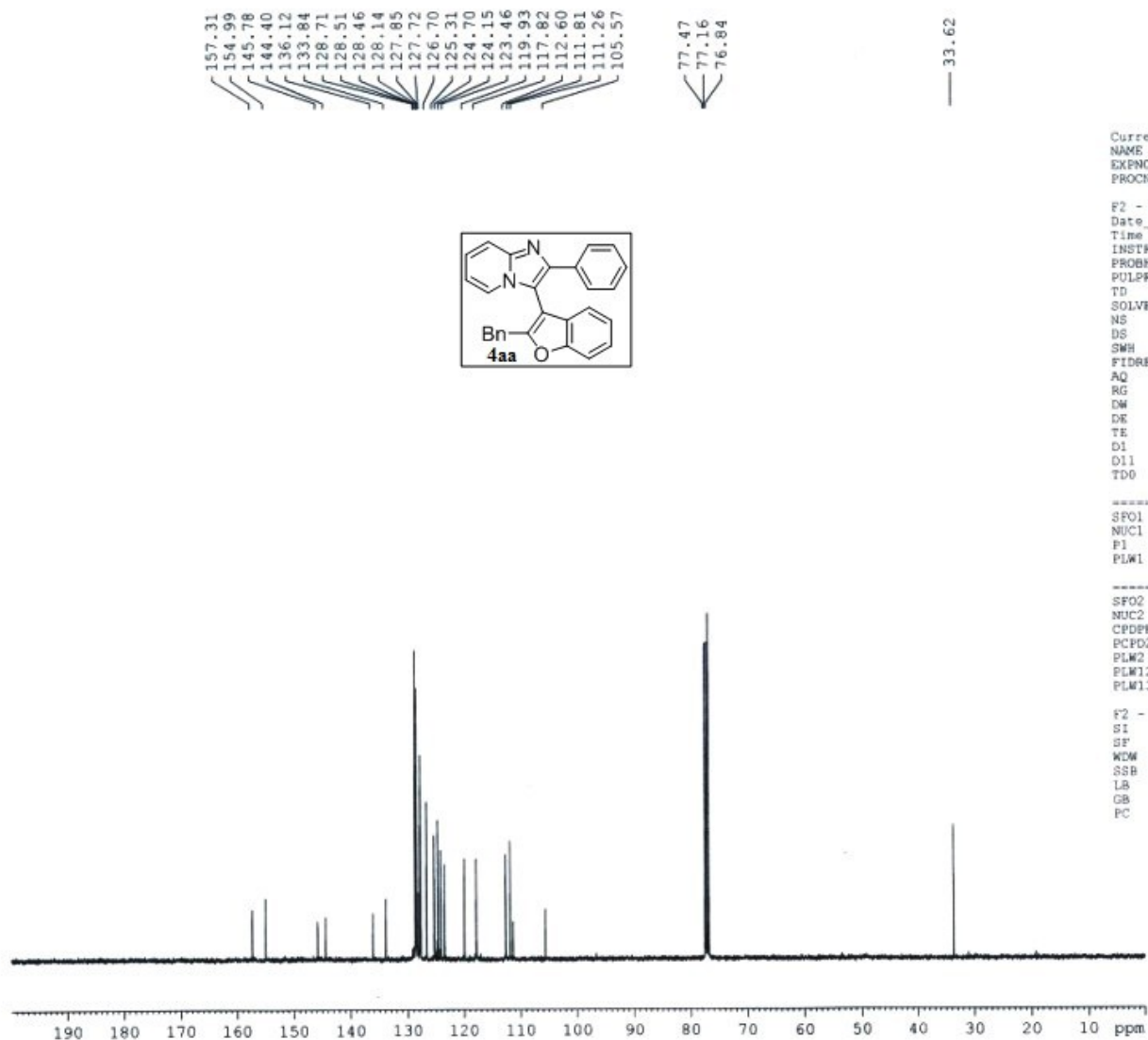
2-(2-Benzylidene-2,3-dihydrobenzofuran-3-yl)phenol (12): Yellow solid (76%, 53 mg); R_f = 0.55 (PE : EA = 94 : 6); ^1H NMR (400 MHz, CDCl_3): δ 7.87 (d, J = 8.4 Hz, 1H), 7.70 (d, J = 8.8 Hz, 1H), 7.63-7.61 (m, 2H), 7.45-7.40 (m, 2H), 7.38-7.25 (m, 4H), 7.22-7.19 (m, 3H), 7.12-7.05 (m, 2H), 5.00 (s, 1H), 4.07-3.98 (m, 2H); $^{13}\text{C}\{^1\text{H}\}$ NMR (100 MHz, CDCl_3): δ 154.8, 154.4, 152.4, 137.6, 131.7, 130.8, 130.4, 128.8, 128.7, 128.0, 126.8, 126.5, 125.7, 124.6, 122.7, 121.9, 121.1, 119.4, 115.8, 112.7, 112.4, 23.0; Anal. Calcd for $\text{C}_{25}\text{H}_{18}\text{O}_2$: C, 85.69; H, 5.18%; Found: C, 85.49; H, 5.22%.

6. References:

- (1) (a) A. K. Bagdi, M. Rahman, S. Santra, A. Majee and A. Hajra, *Adv. Synth. Catal.*, 2013, **355**, 1741;
 (b) S. Mishra, K. Monir, S. Mitra and A. Hajra, *Org. Lett.*, 2014, **16**, 6084.
- (2) N. Wongsu, U. Sommart, T. Ritthiwigrom, A. Yazici, S. Kanokmedhakul, K. Kanokmedhakul, A. C. Willis and S. G. Pyne, *J. Org. Chem.*, 2013, **78**, 1138.

7. NMR spectra for the synthesized products





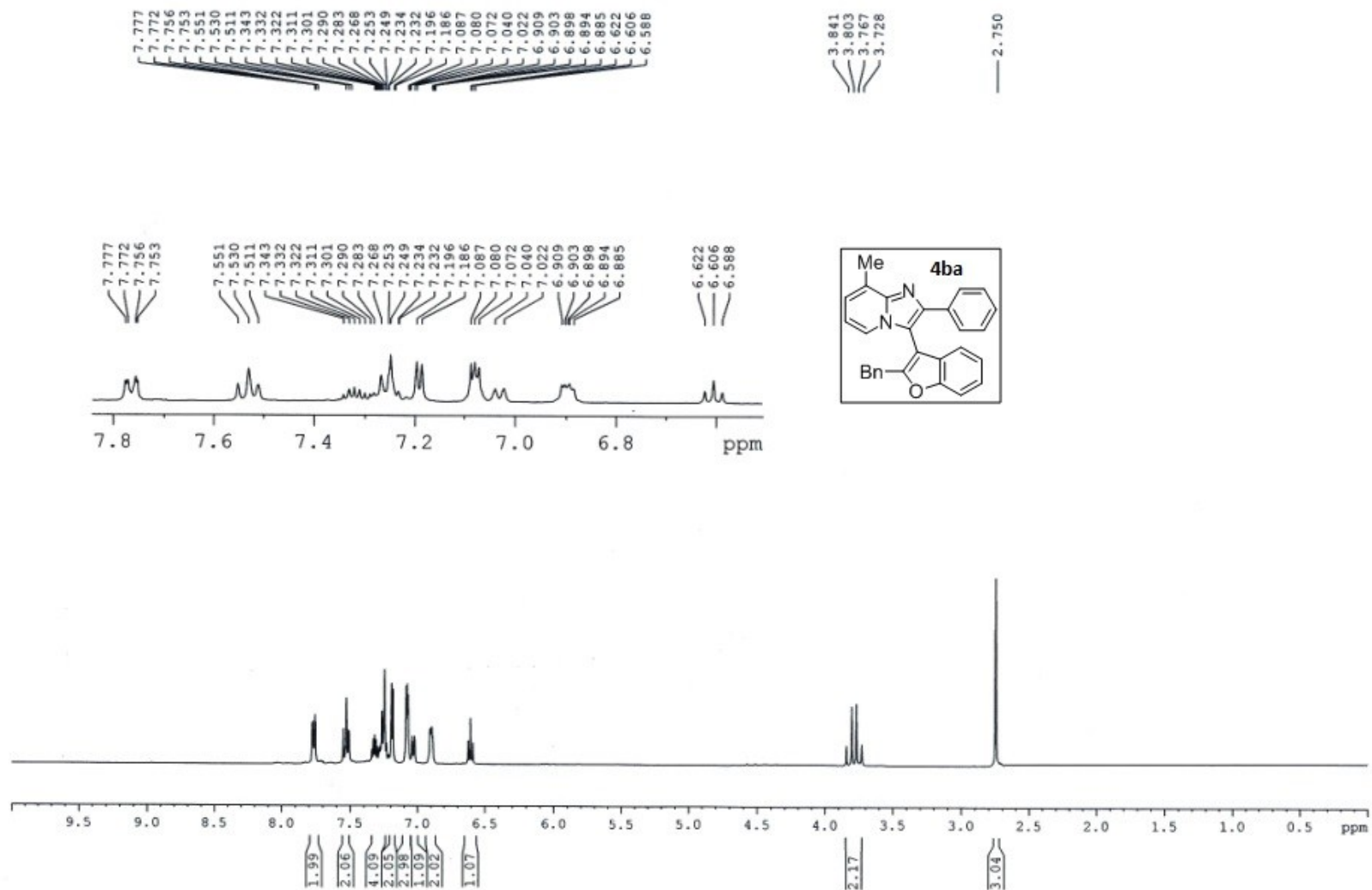
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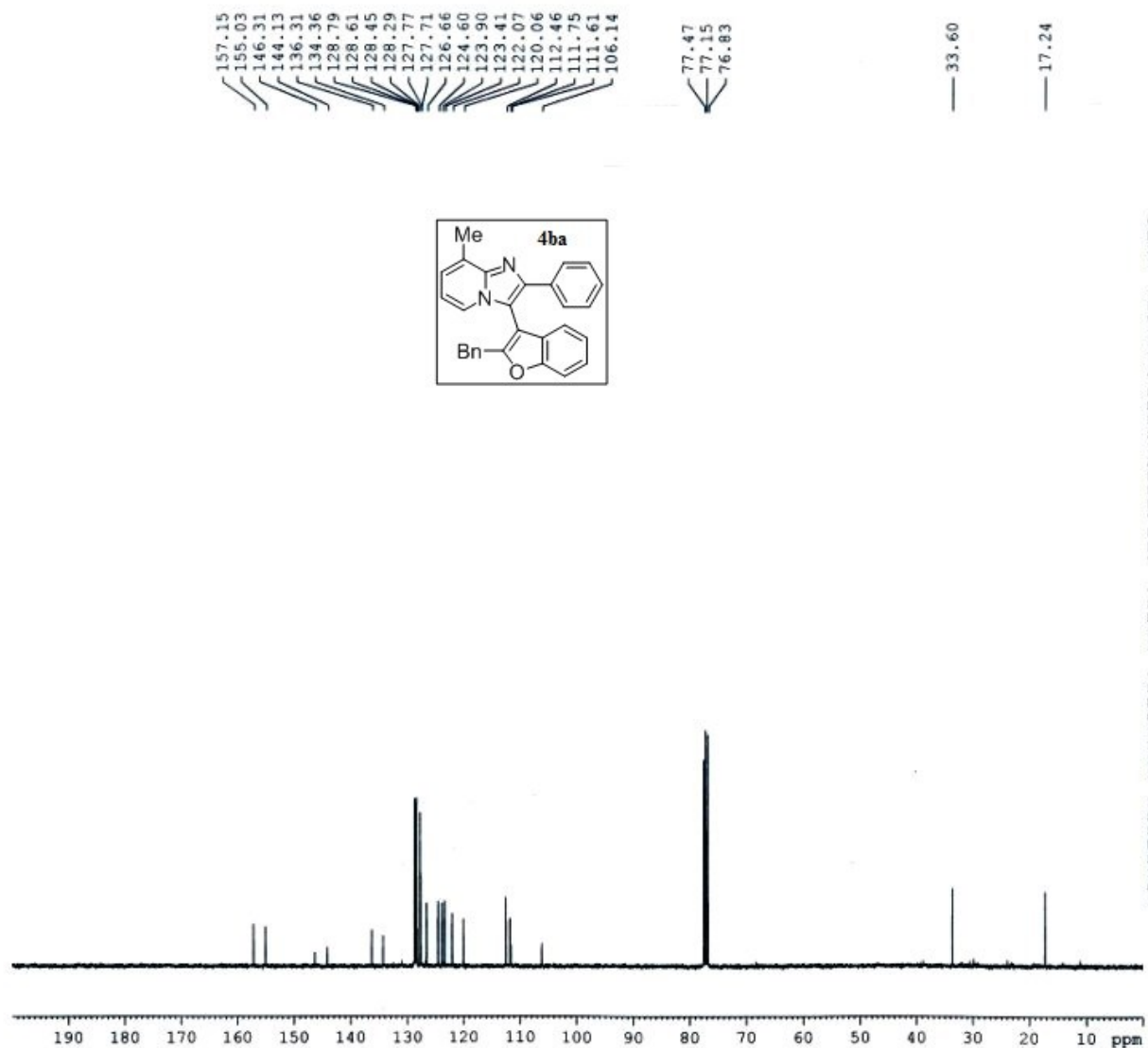
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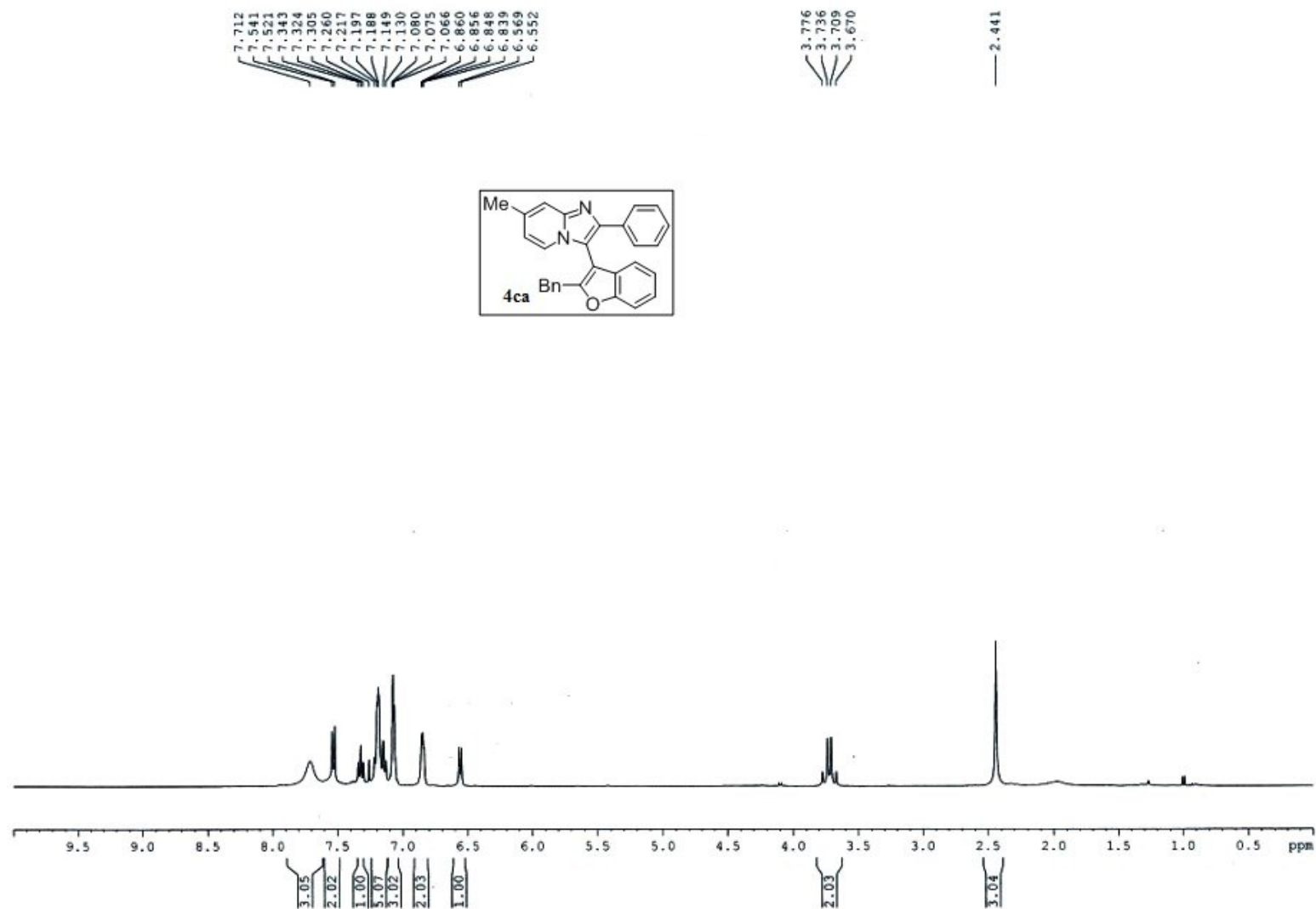
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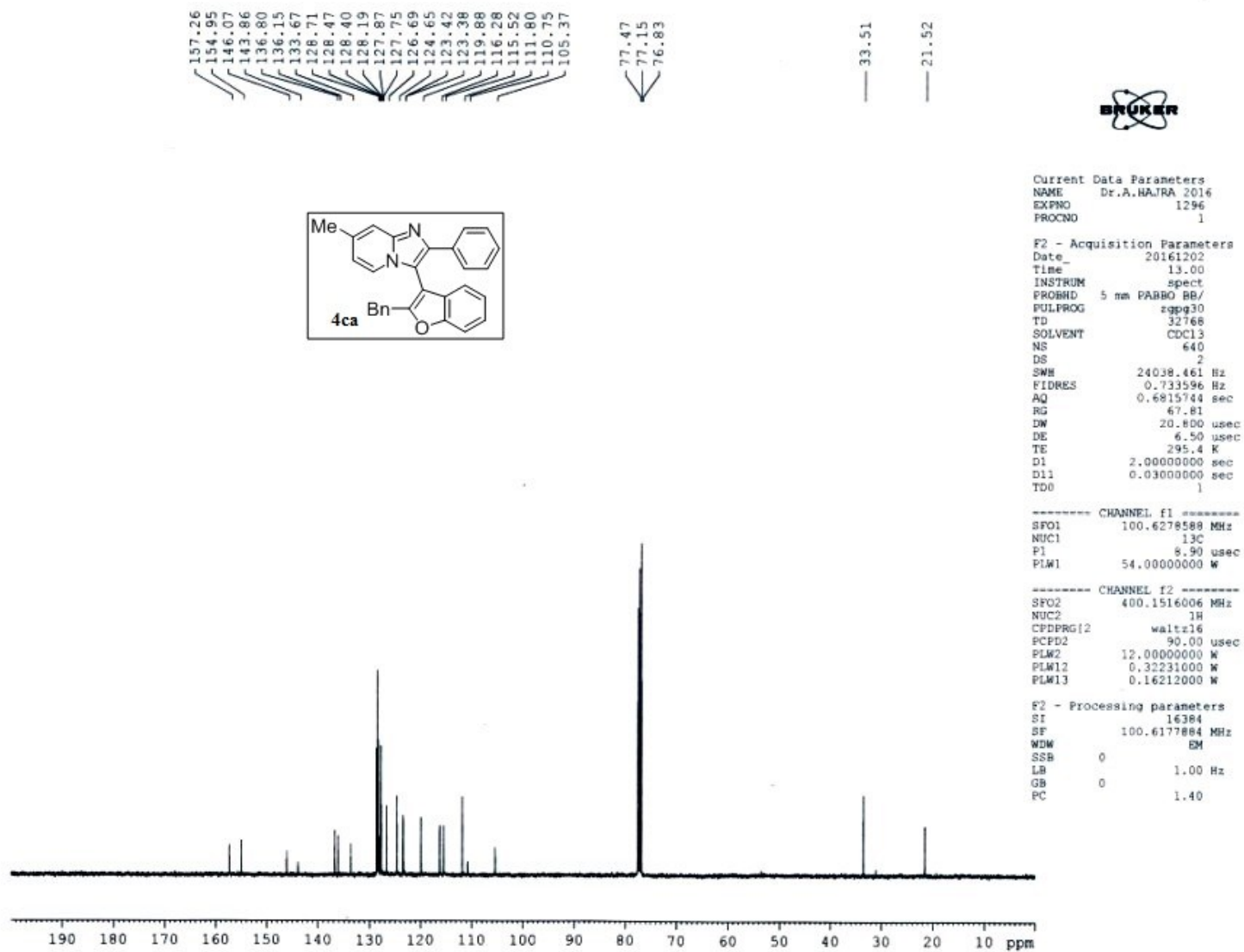
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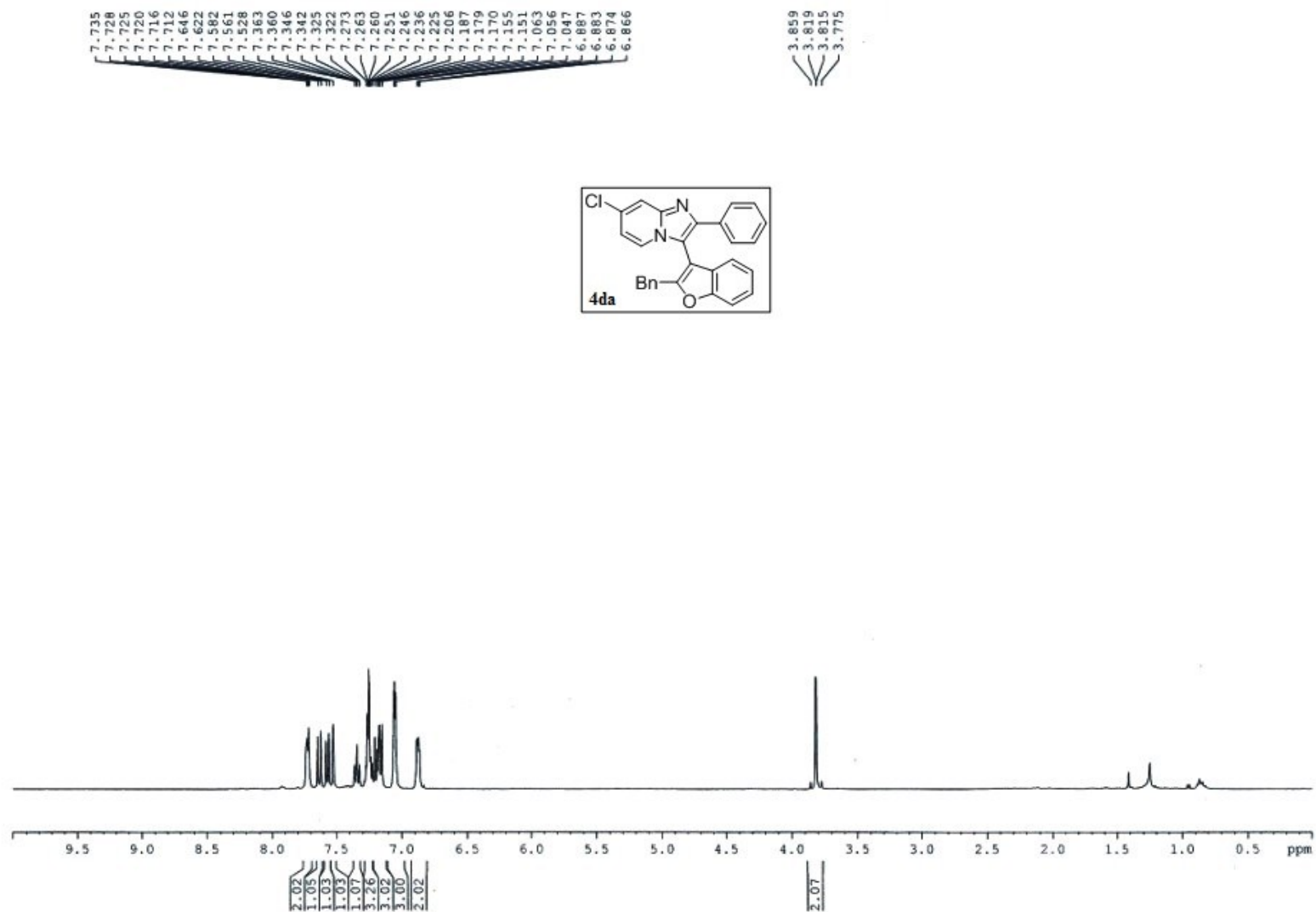
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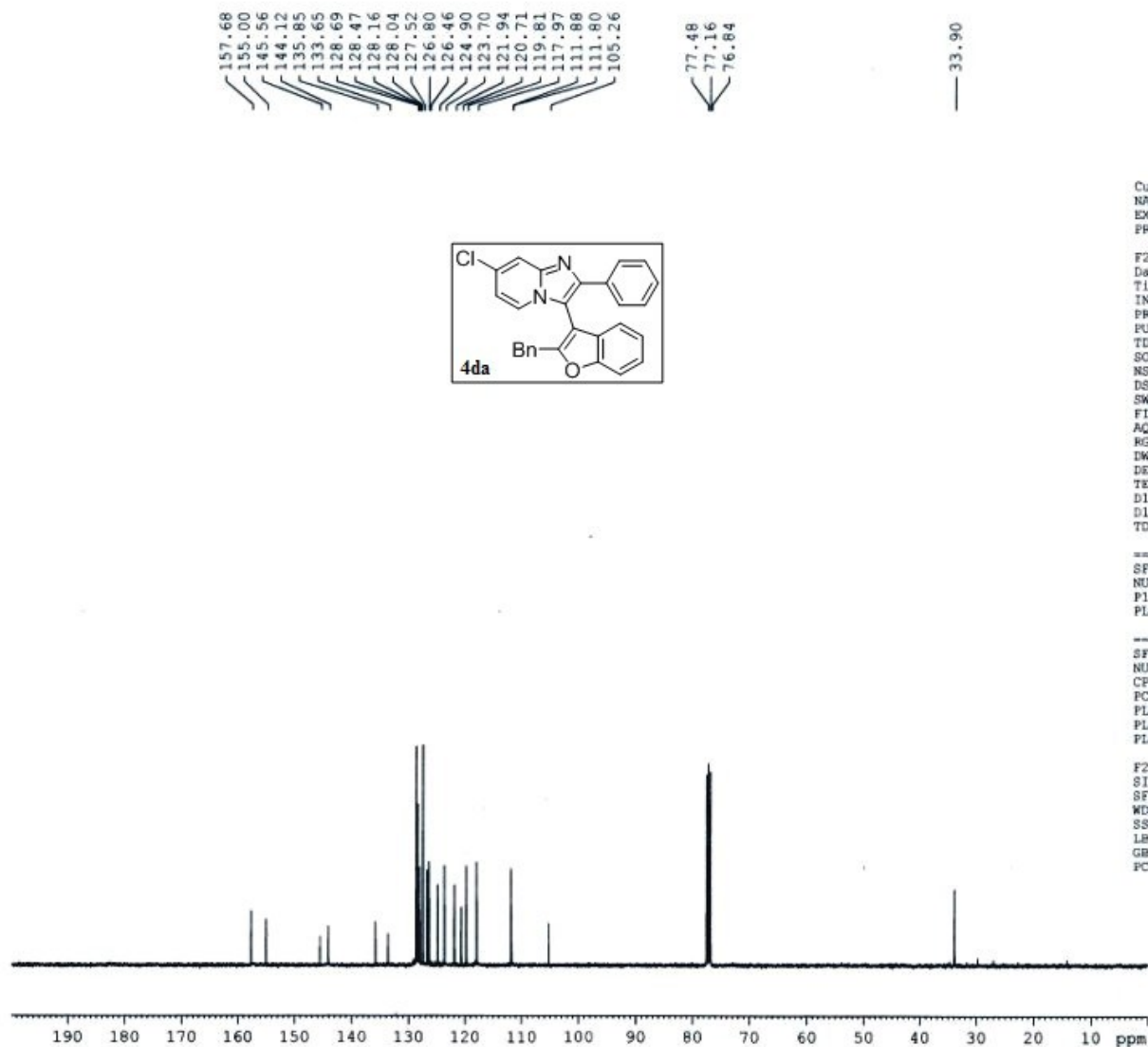
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WDW EM
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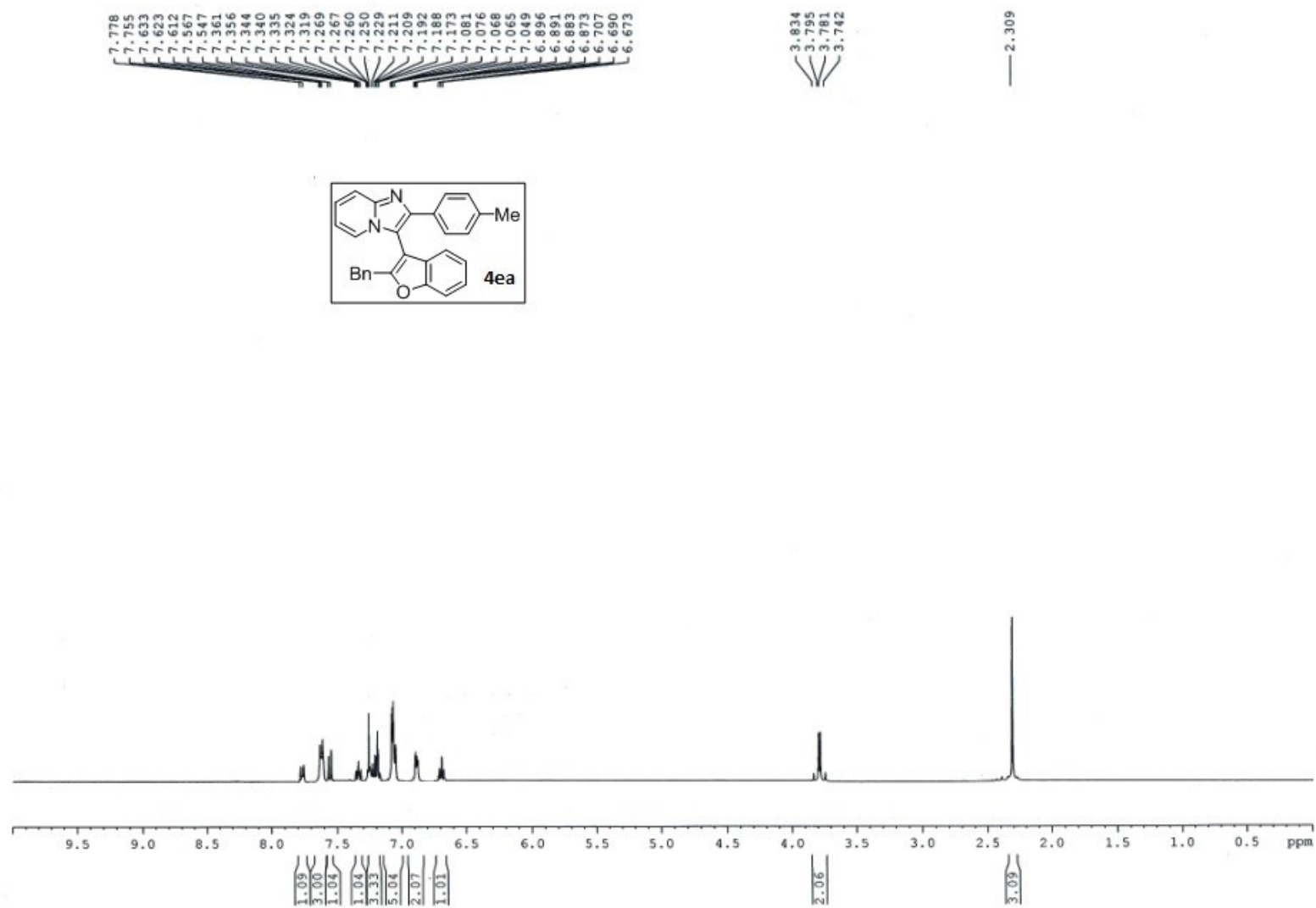
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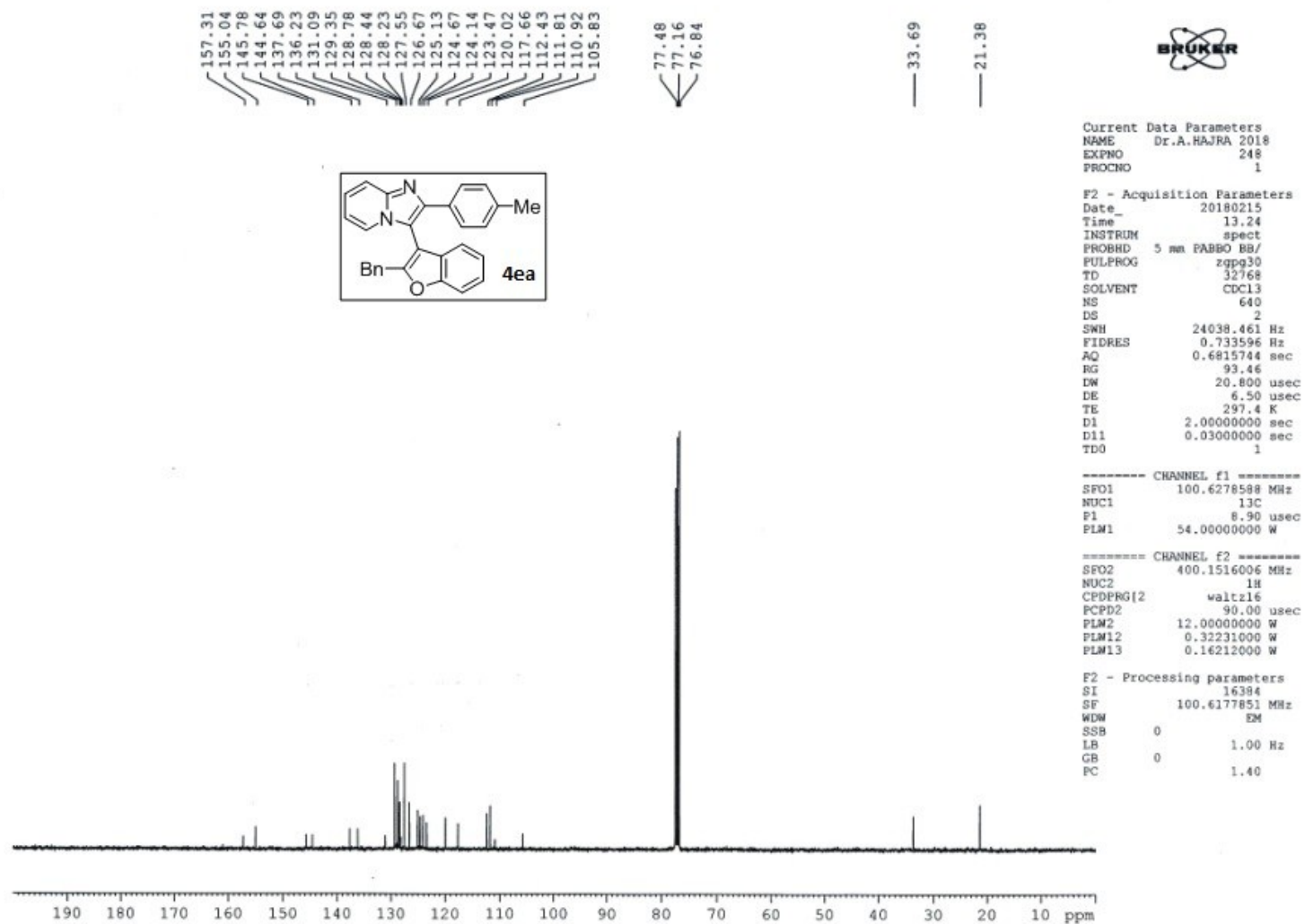
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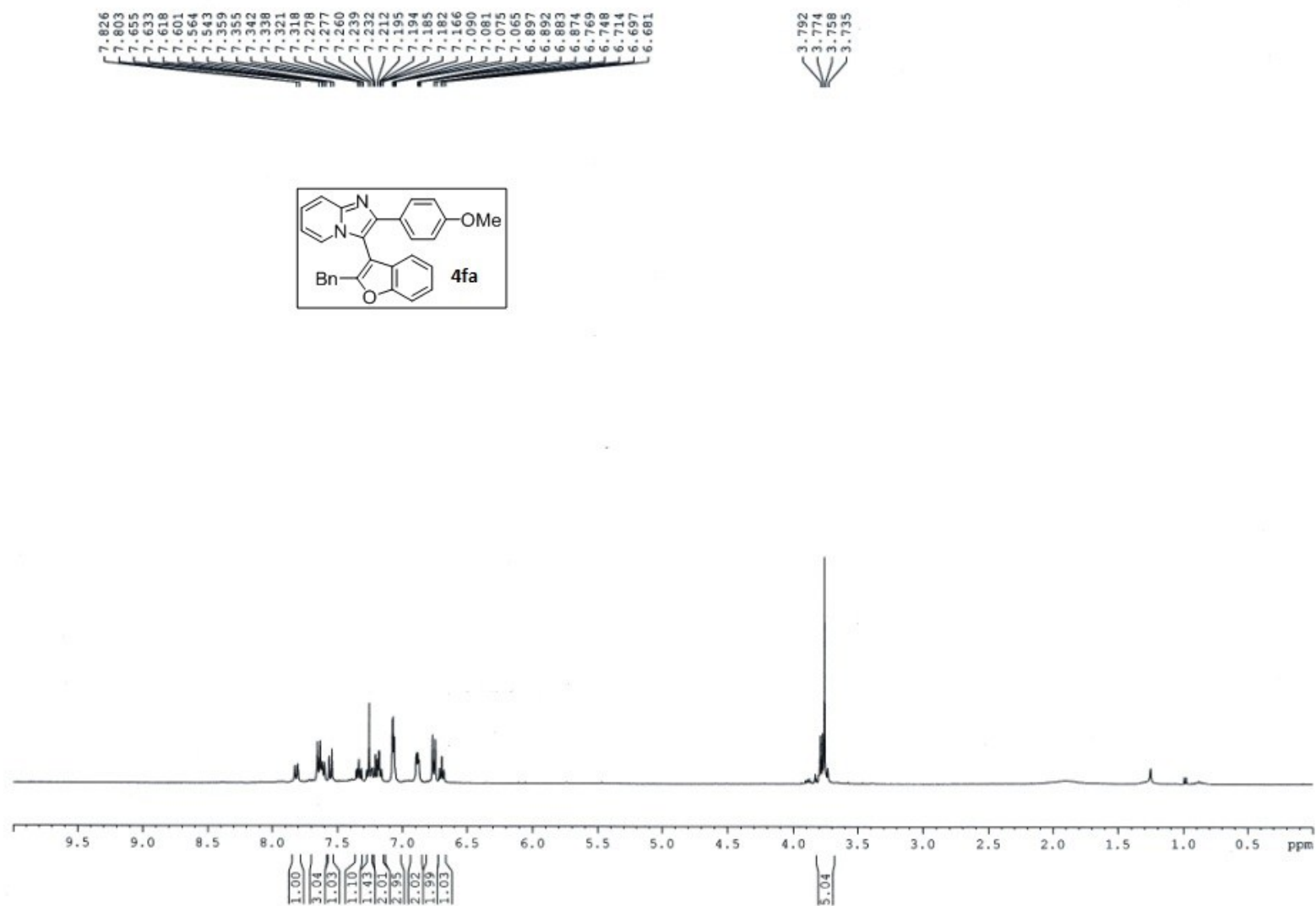
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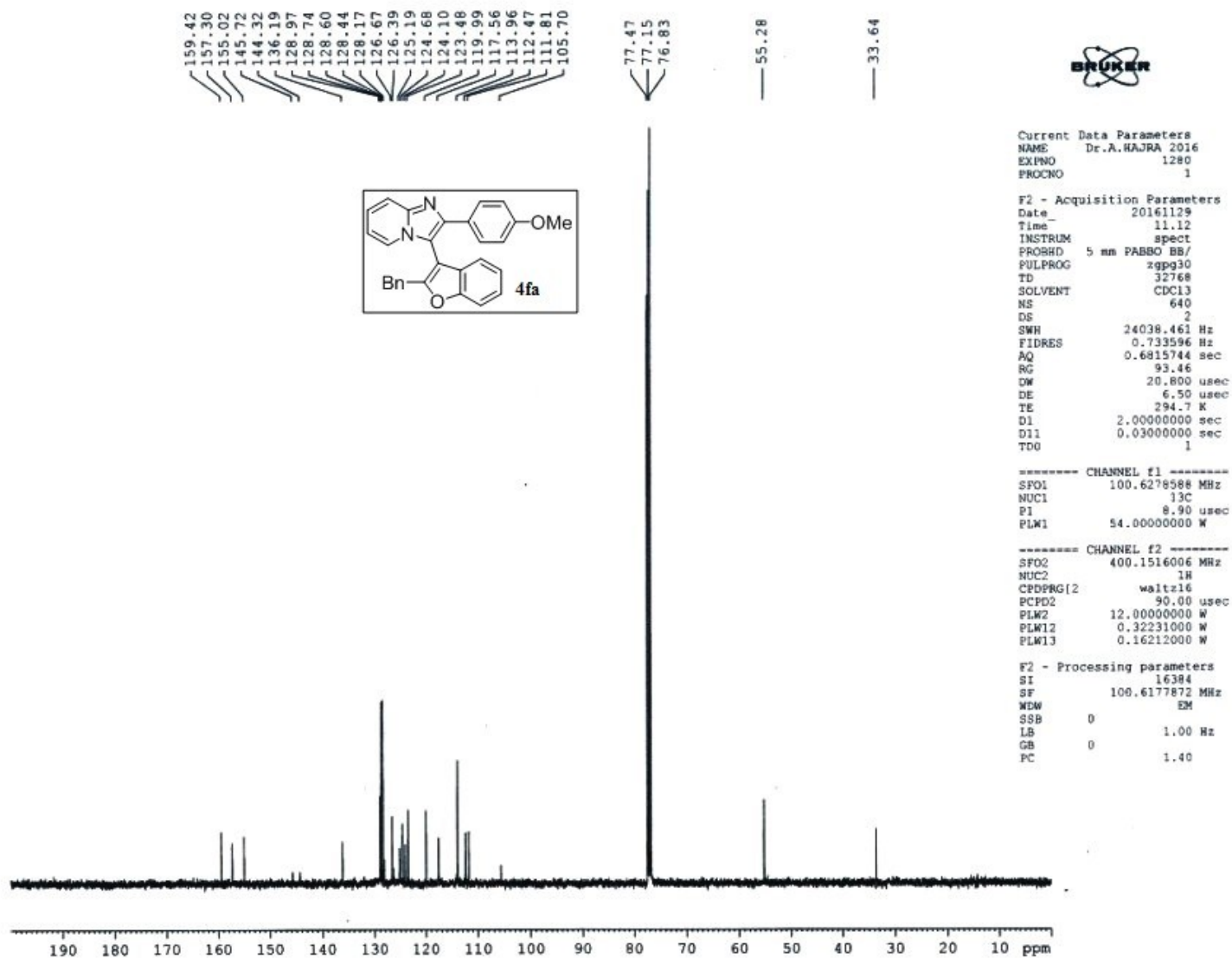
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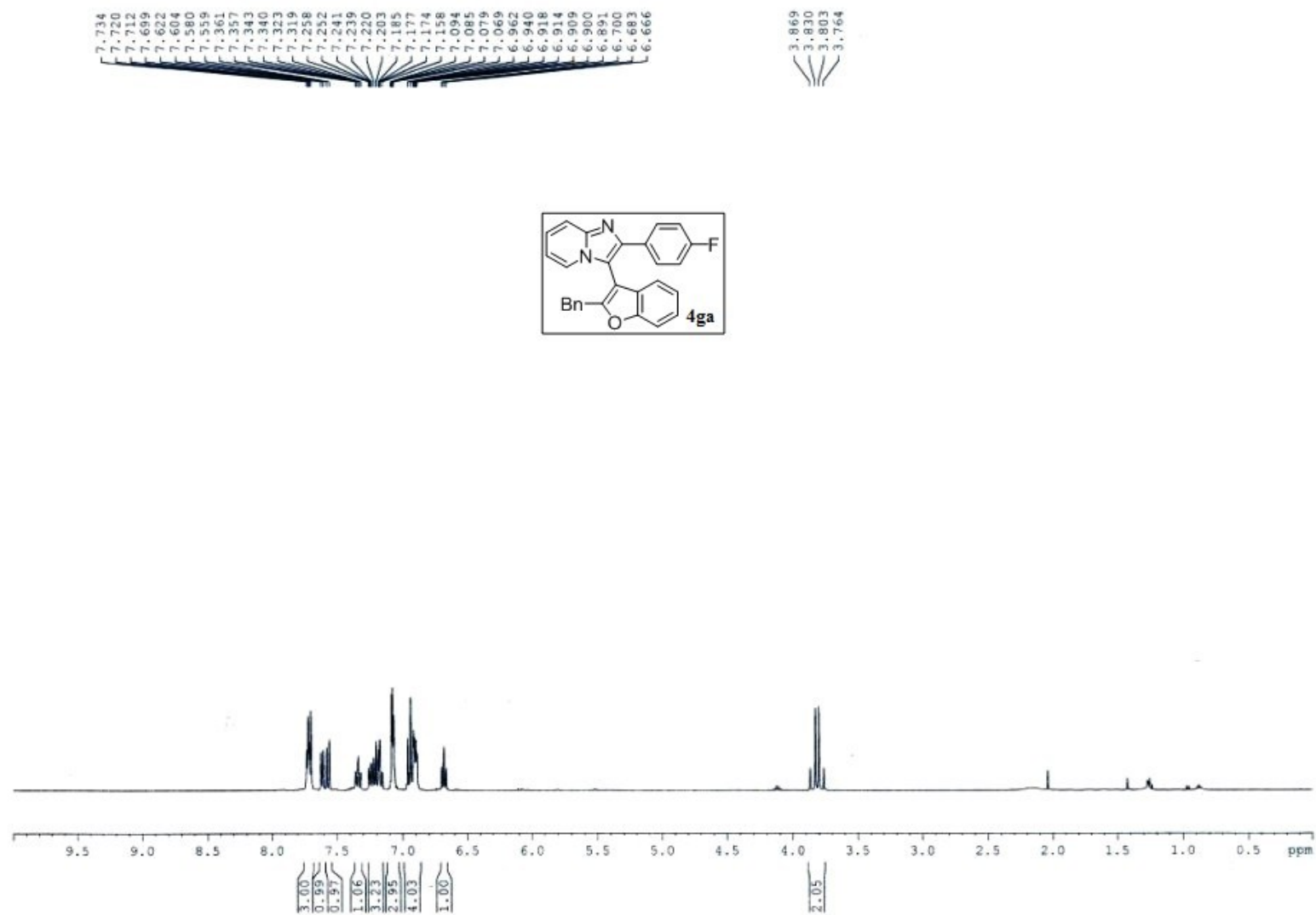
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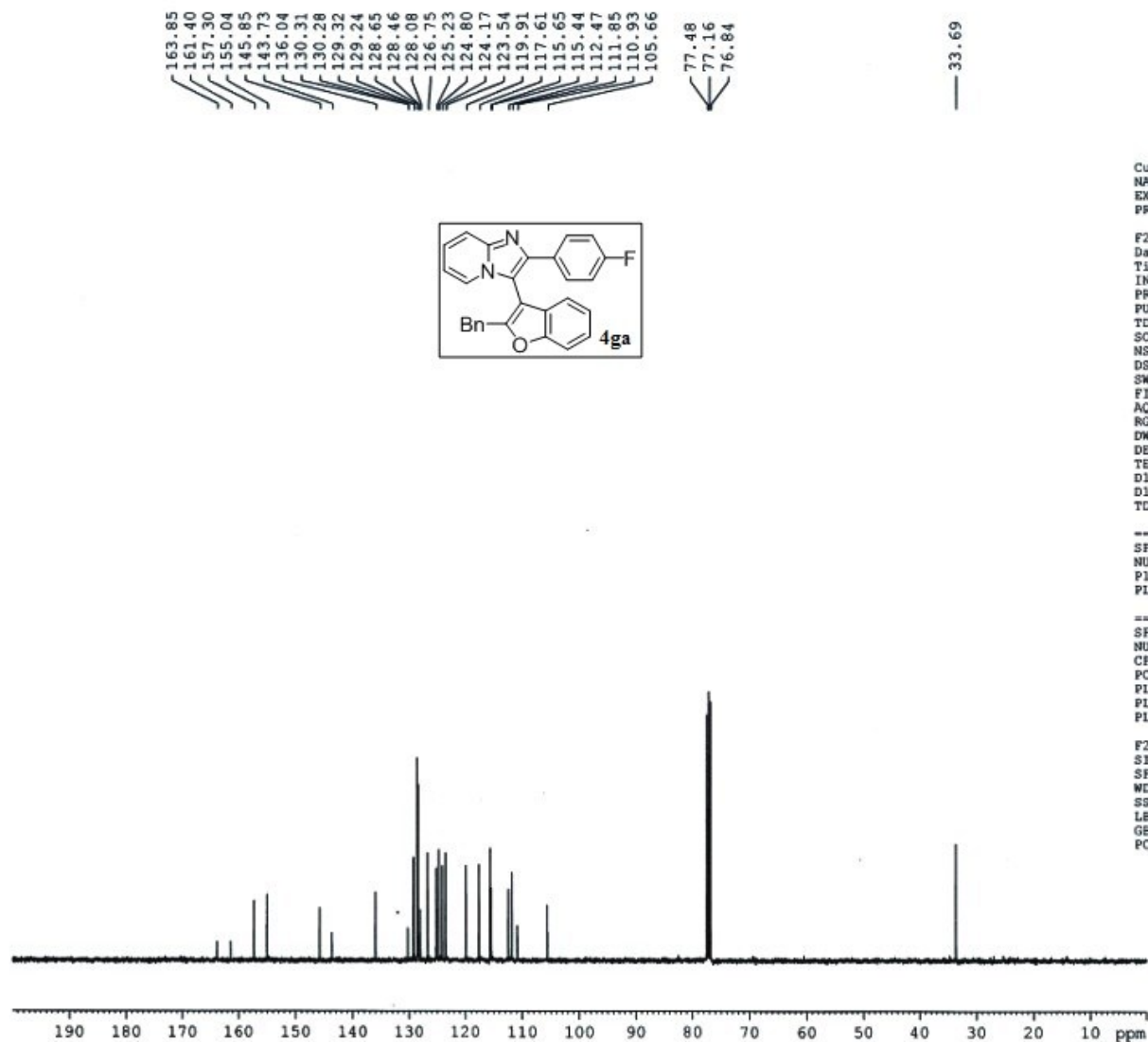












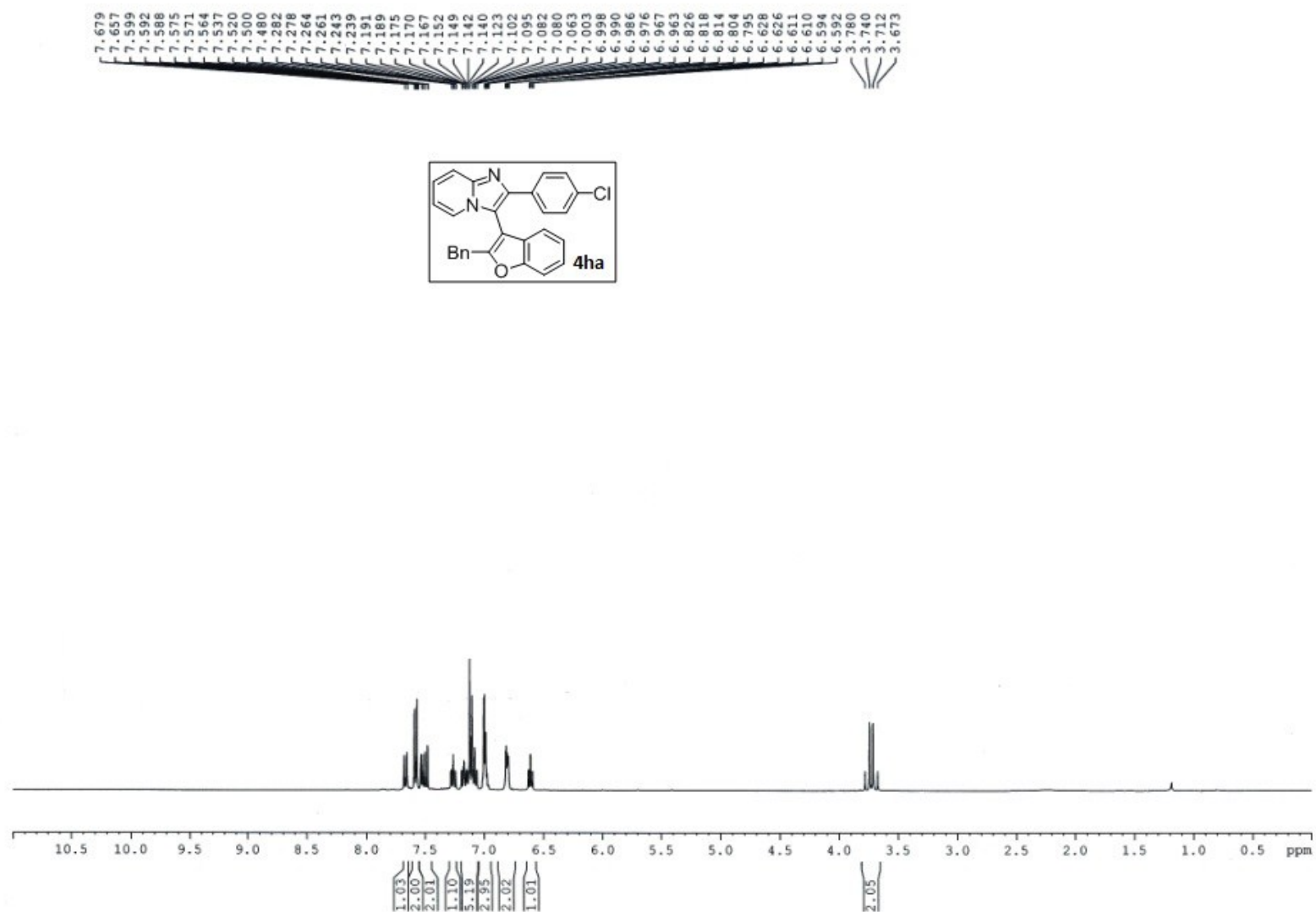
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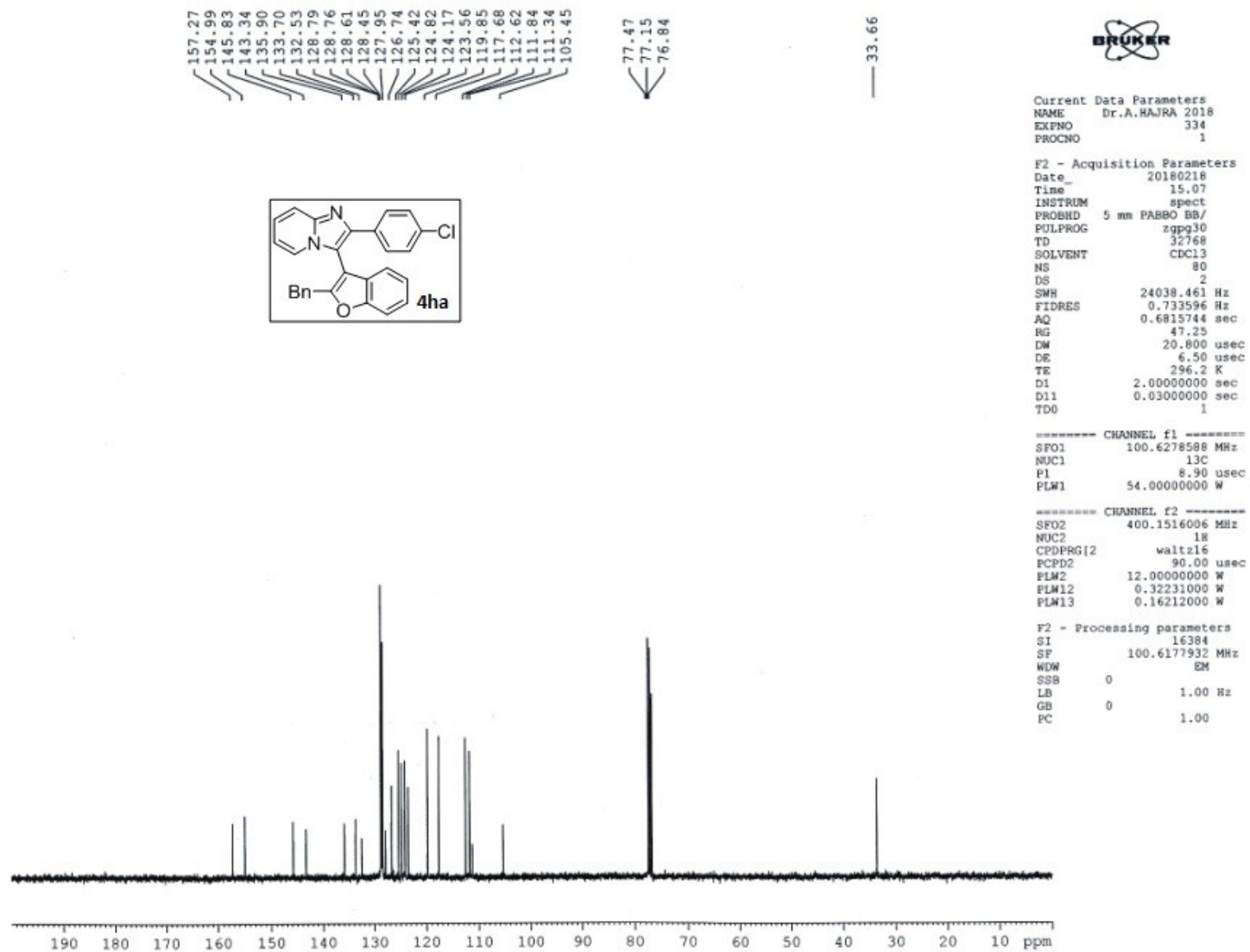
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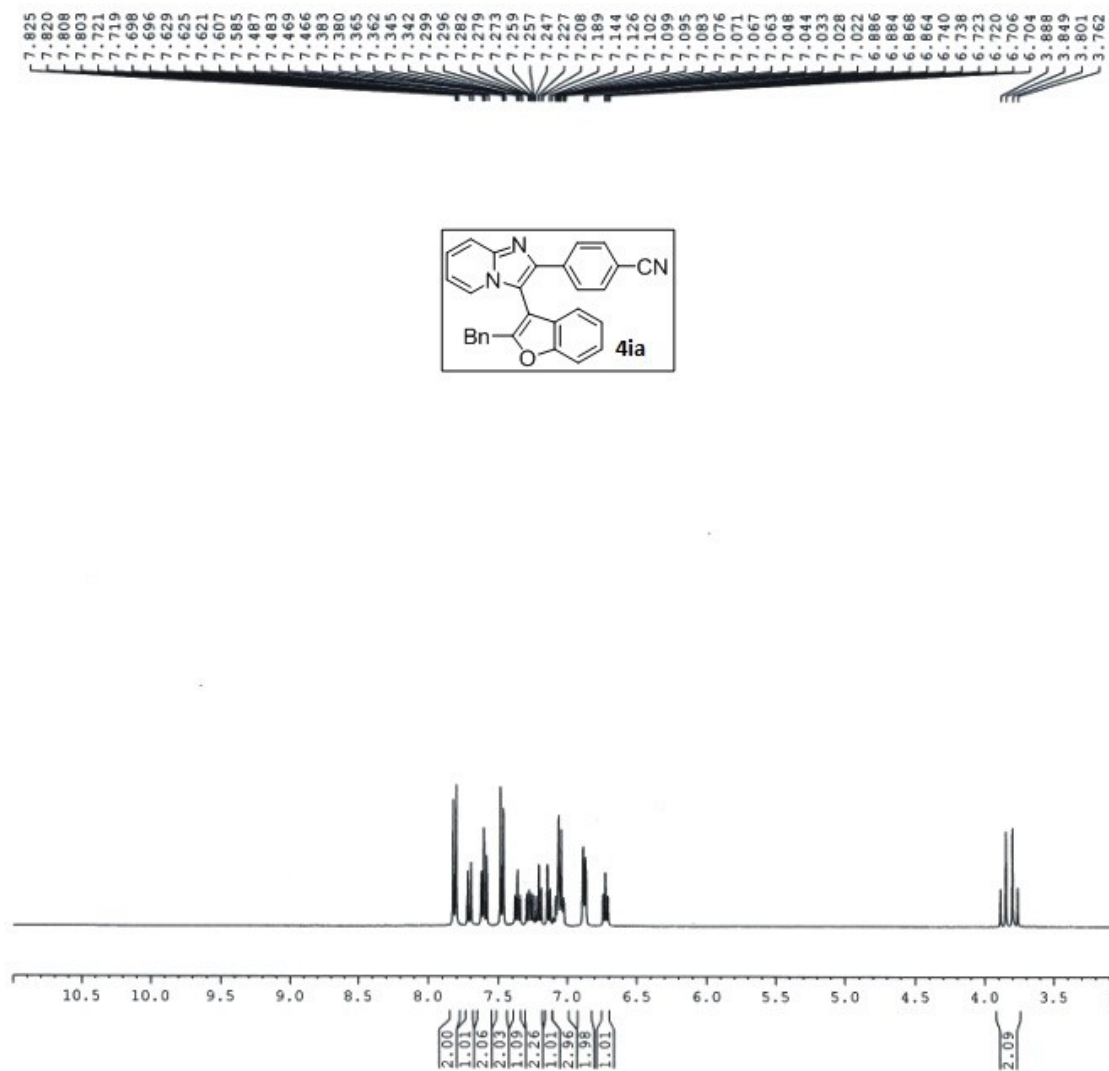
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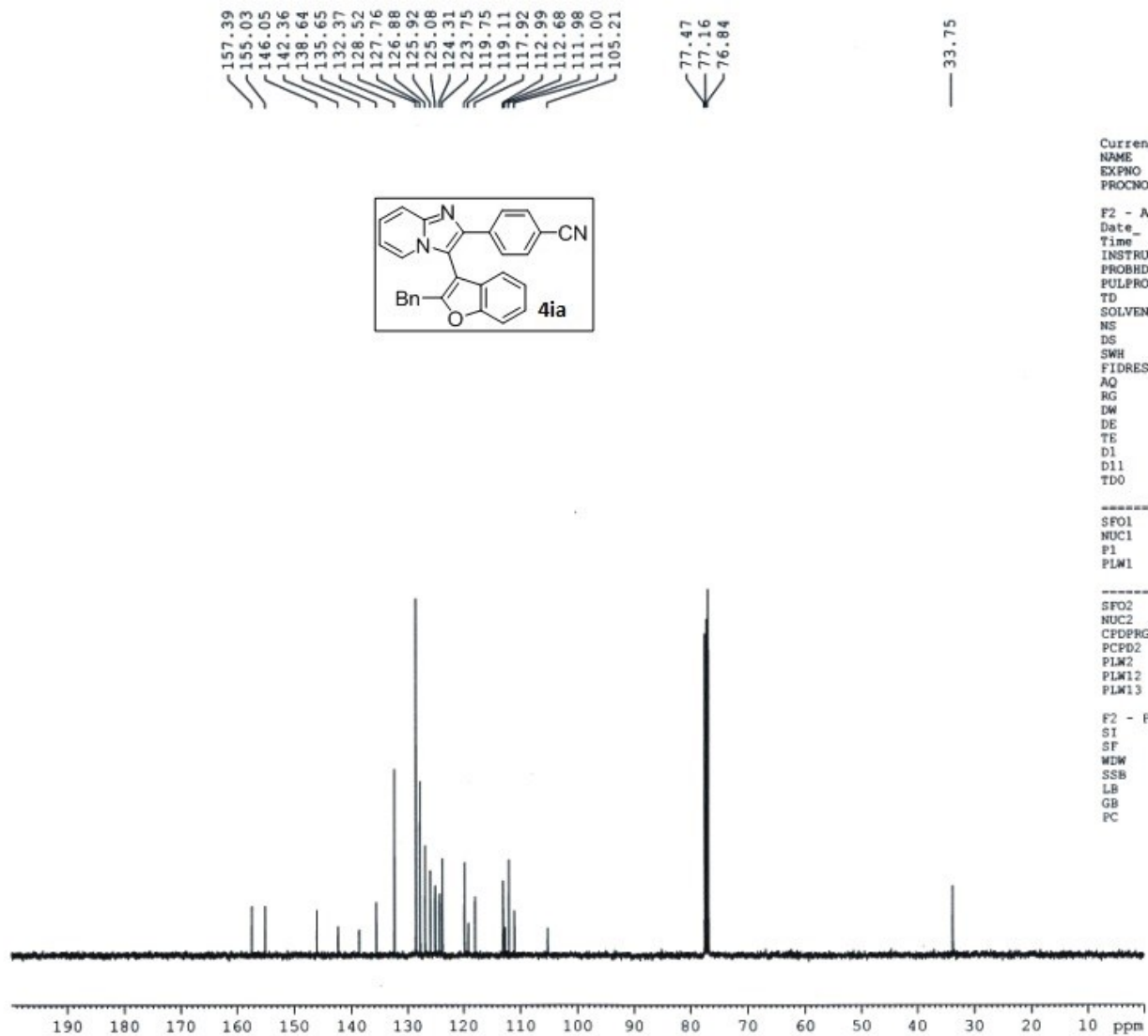
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Current Data Parameters
 NAME Dr.A.HAJRA 2018
 EXPNO 323
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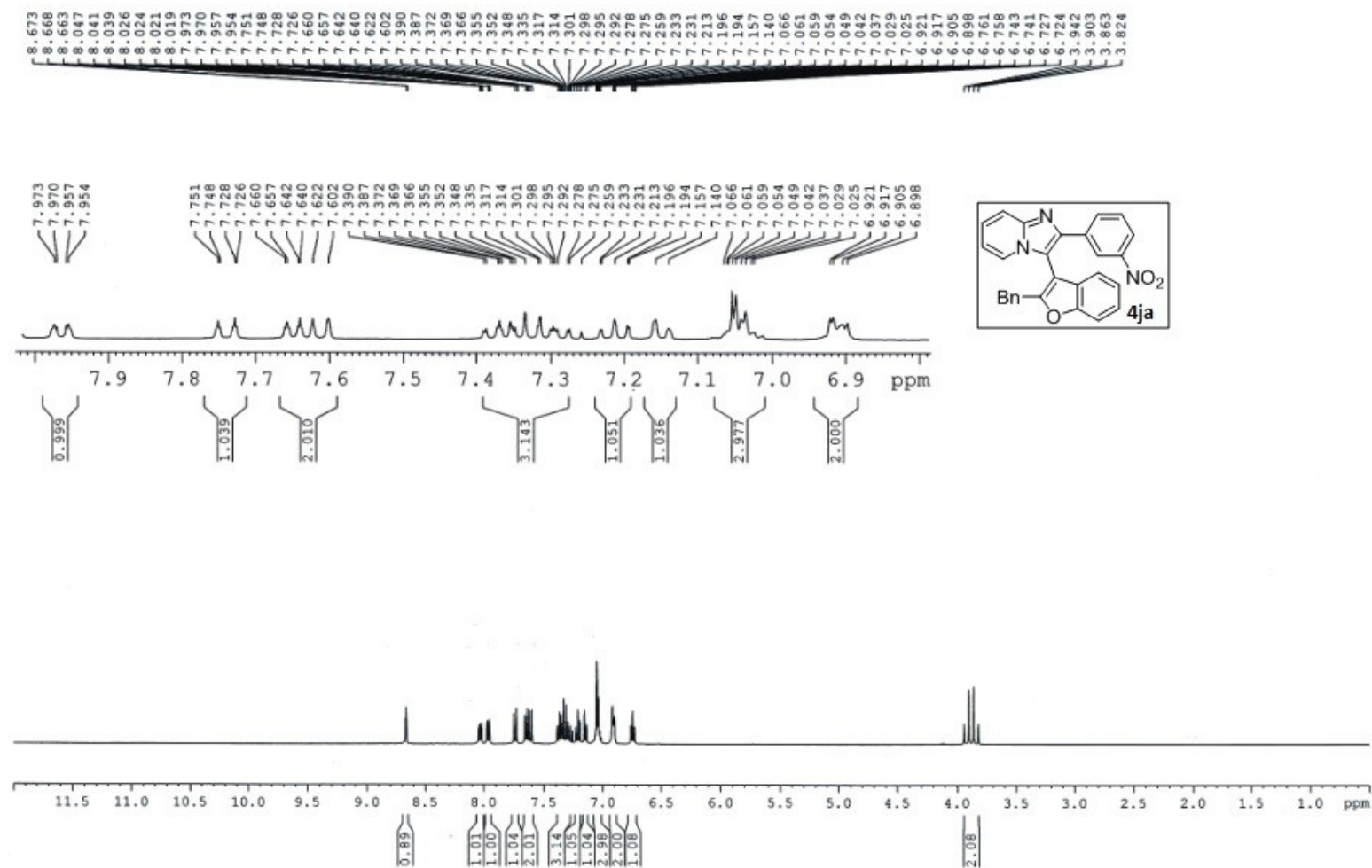
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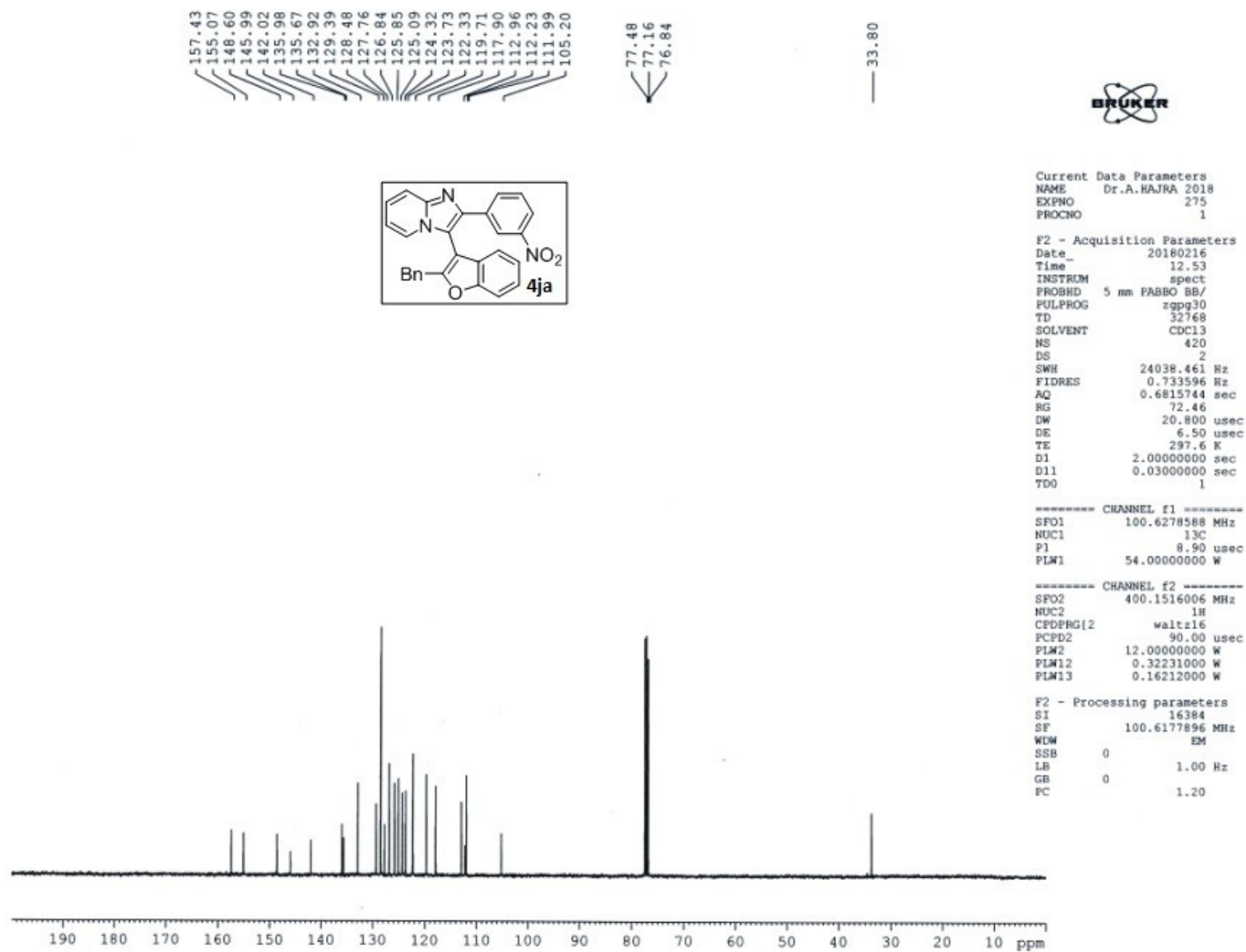
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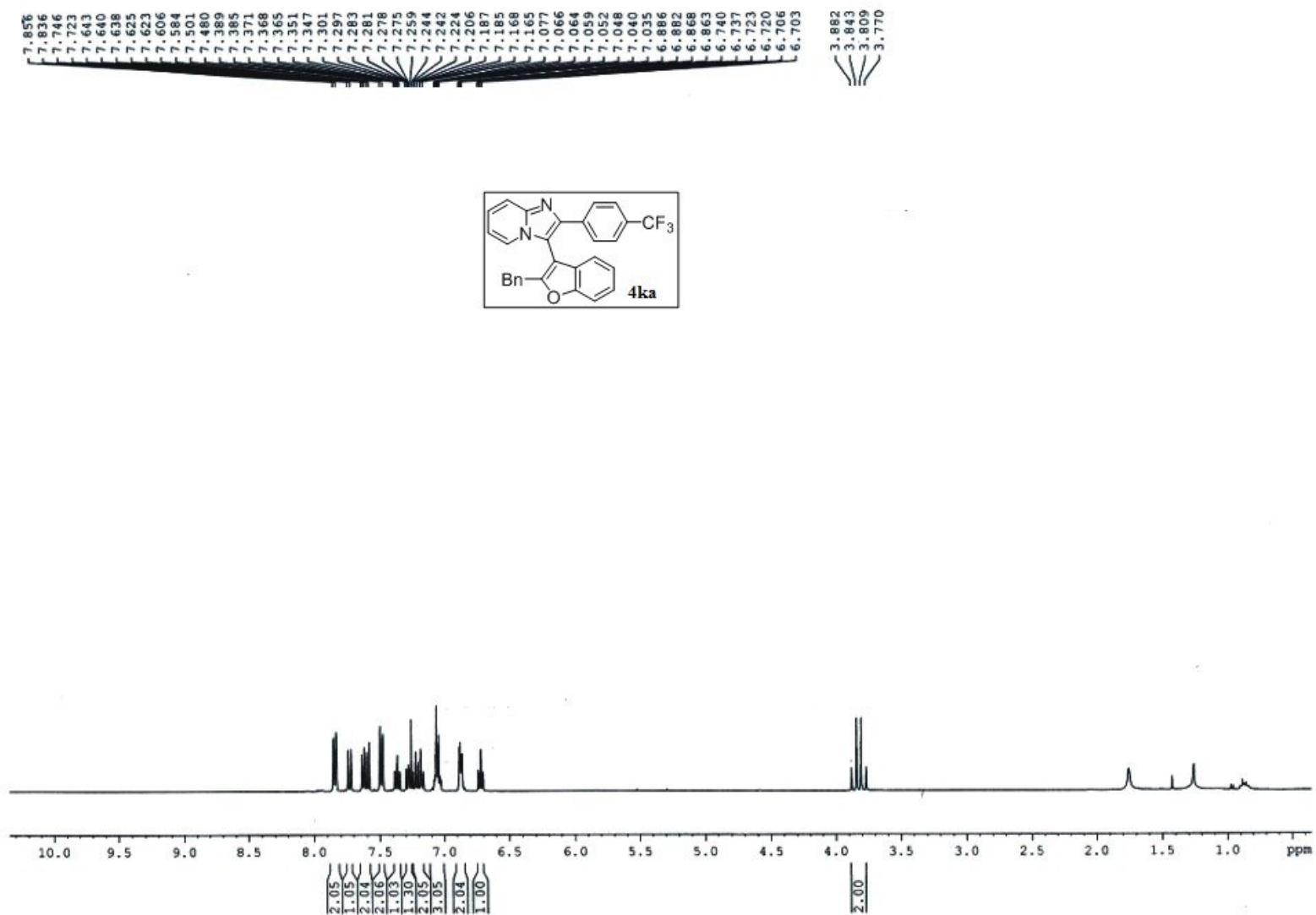
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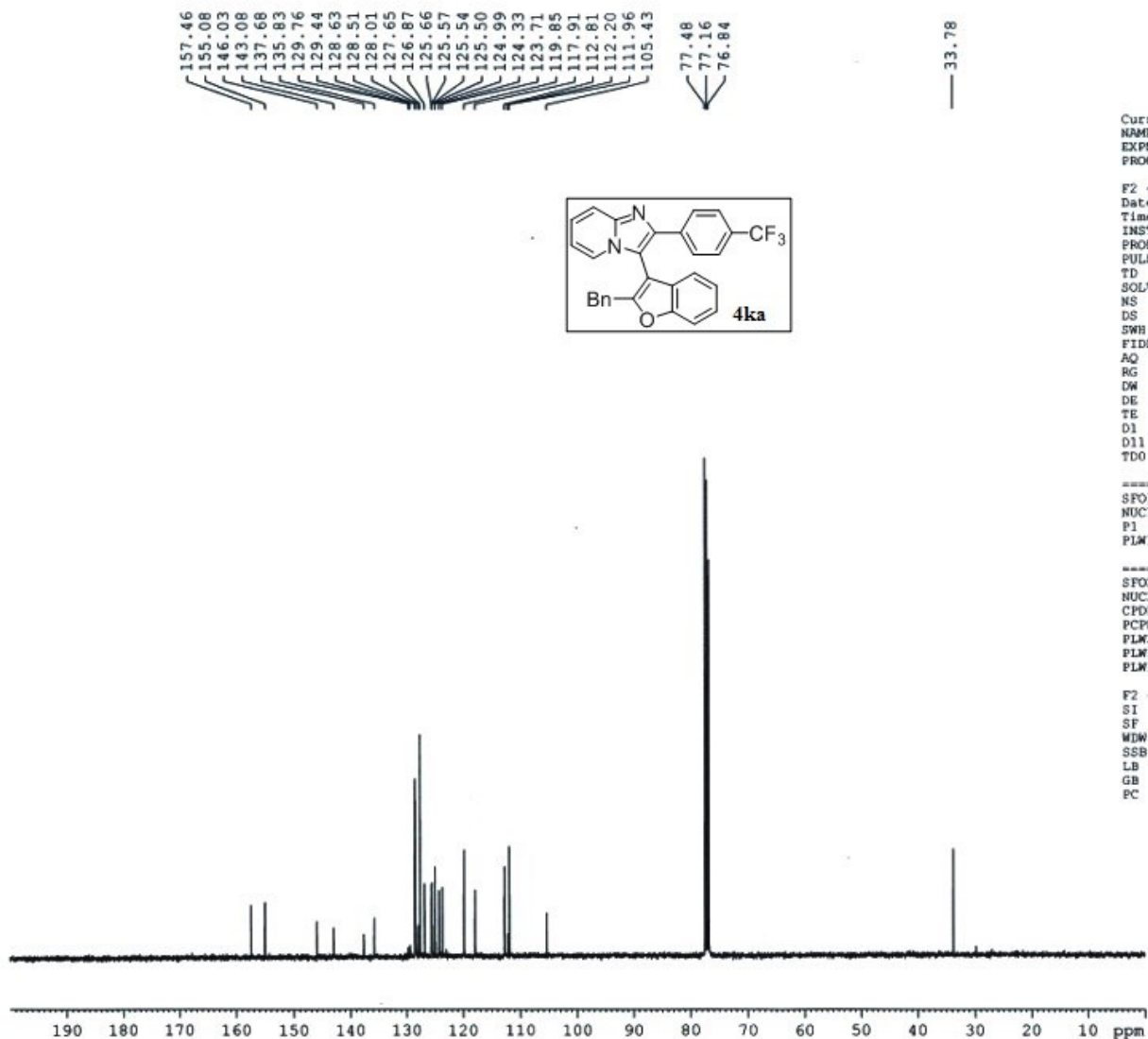
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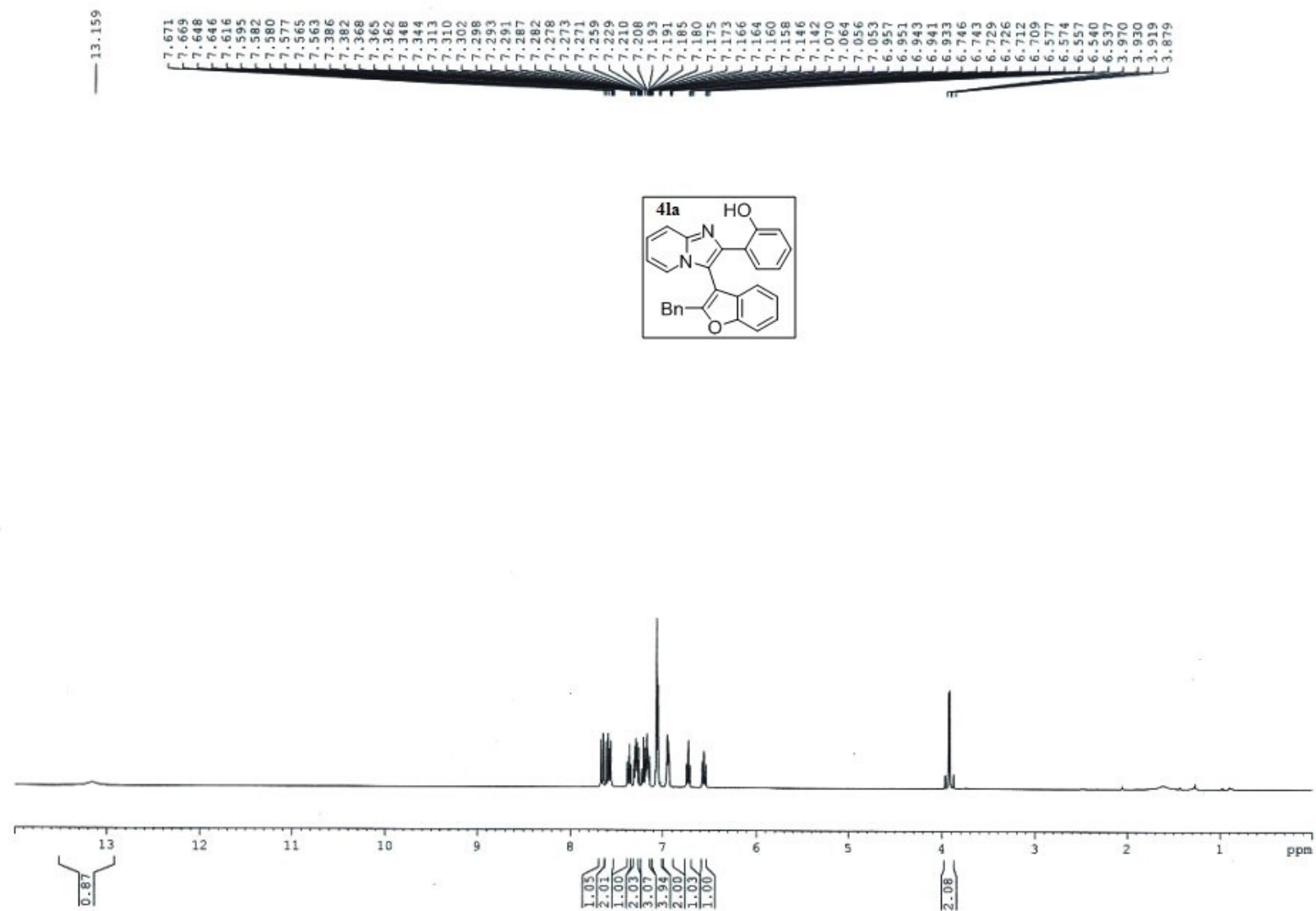
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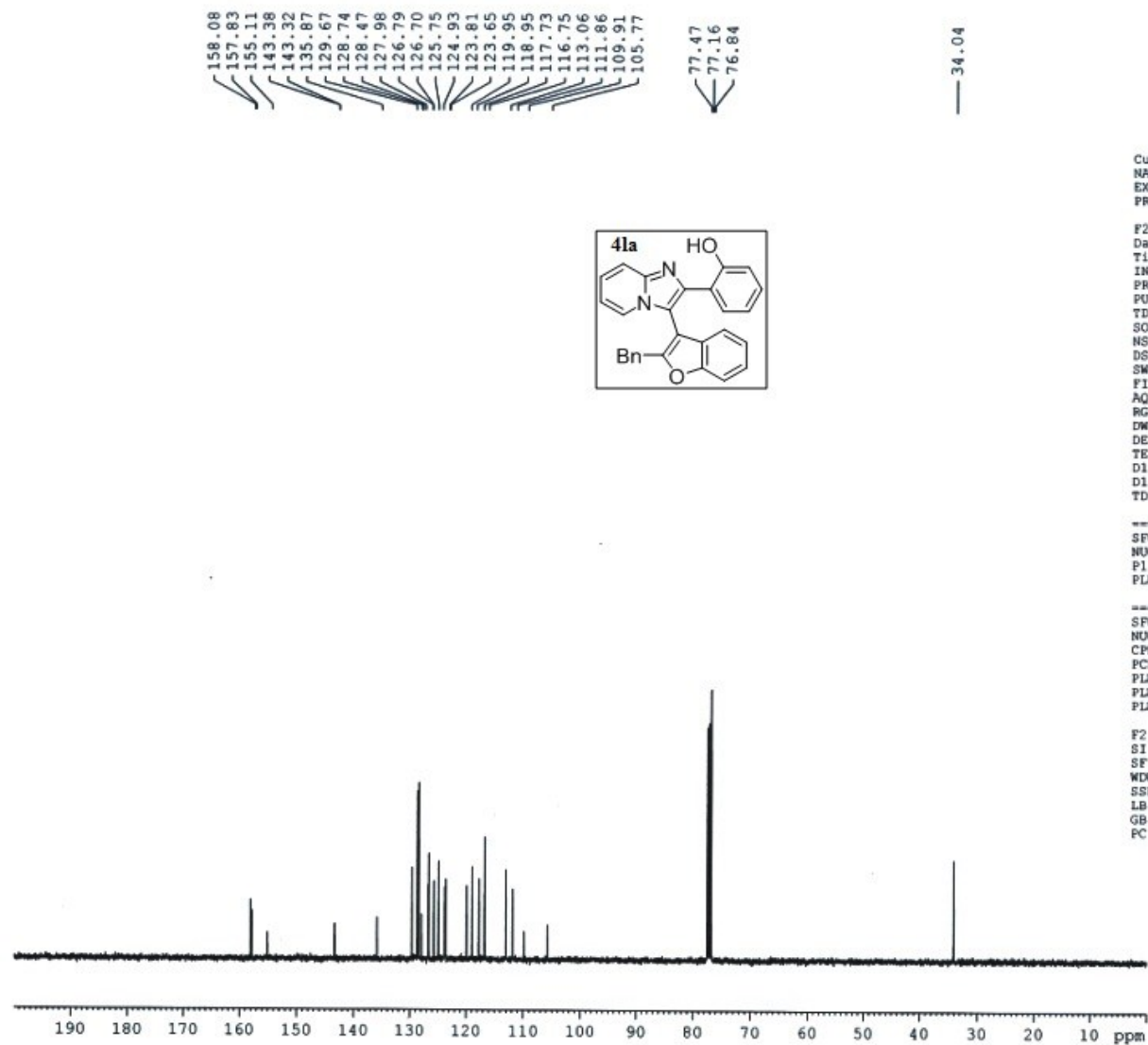
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 AQ 0.6815744 sec
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 D1 2.00000000 sec
 D11 0.03000000 sec
 TDO 1

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===== CHANNEL f2 =====
 SFO2 400.1516006 MHz
 NUC2 1H
 CPDPRG12 waltz16
 PCPD2 90.00 usec
 PLW2 12.00000000 W
 PLW12 0.32231000 W
 PLW13 0.16212000 W

F2 - Processing parameters
 SI 16384
 SF 100.6177838 MHz
 WDW EM
 SSB 0
 LB 1.00 Hz
 GB 0
 PC 1.40





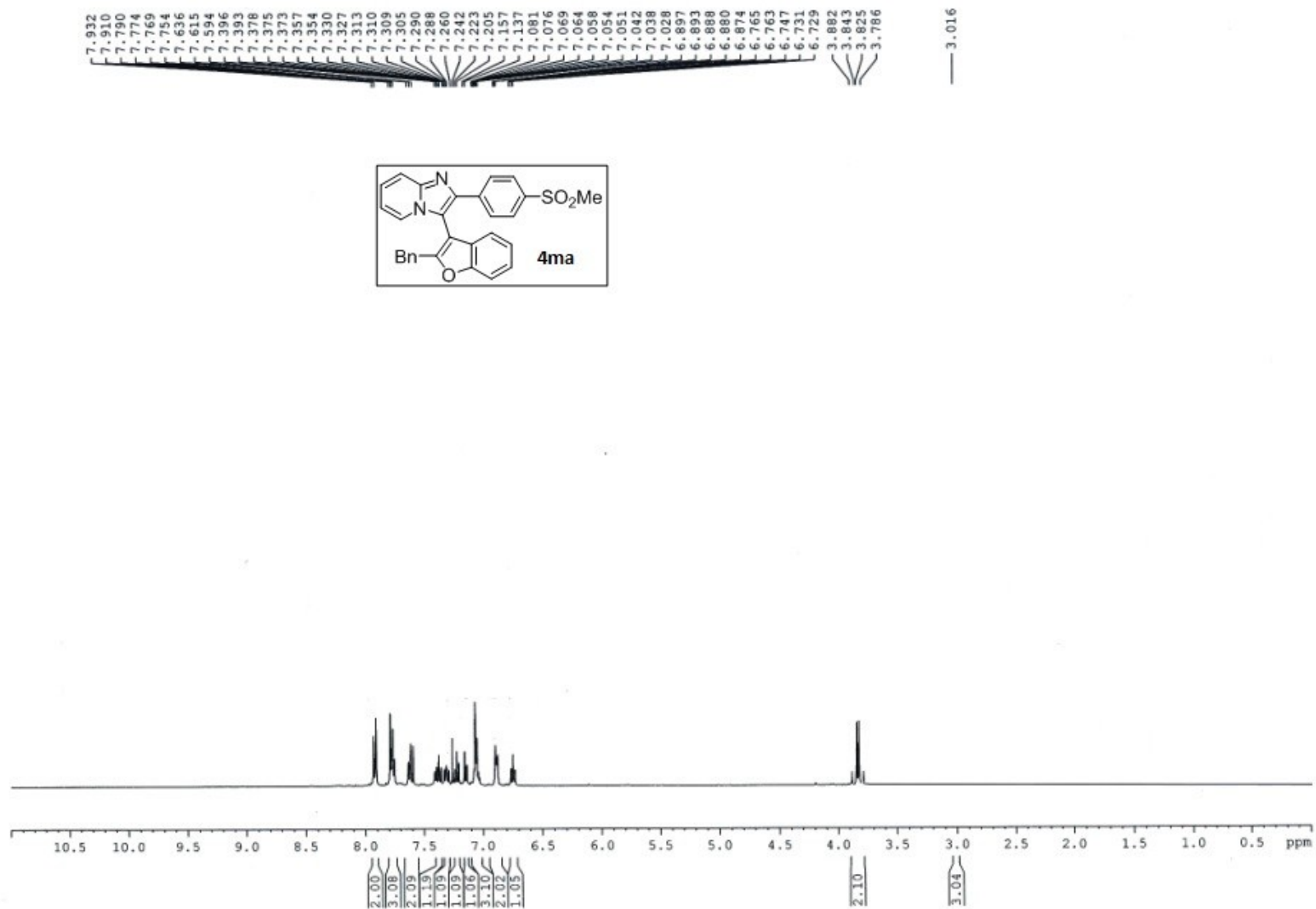
Current Data Parameters
 NAME Dr.A.HAJRA 2017
 EXPNO 1189
 PROCNO 1

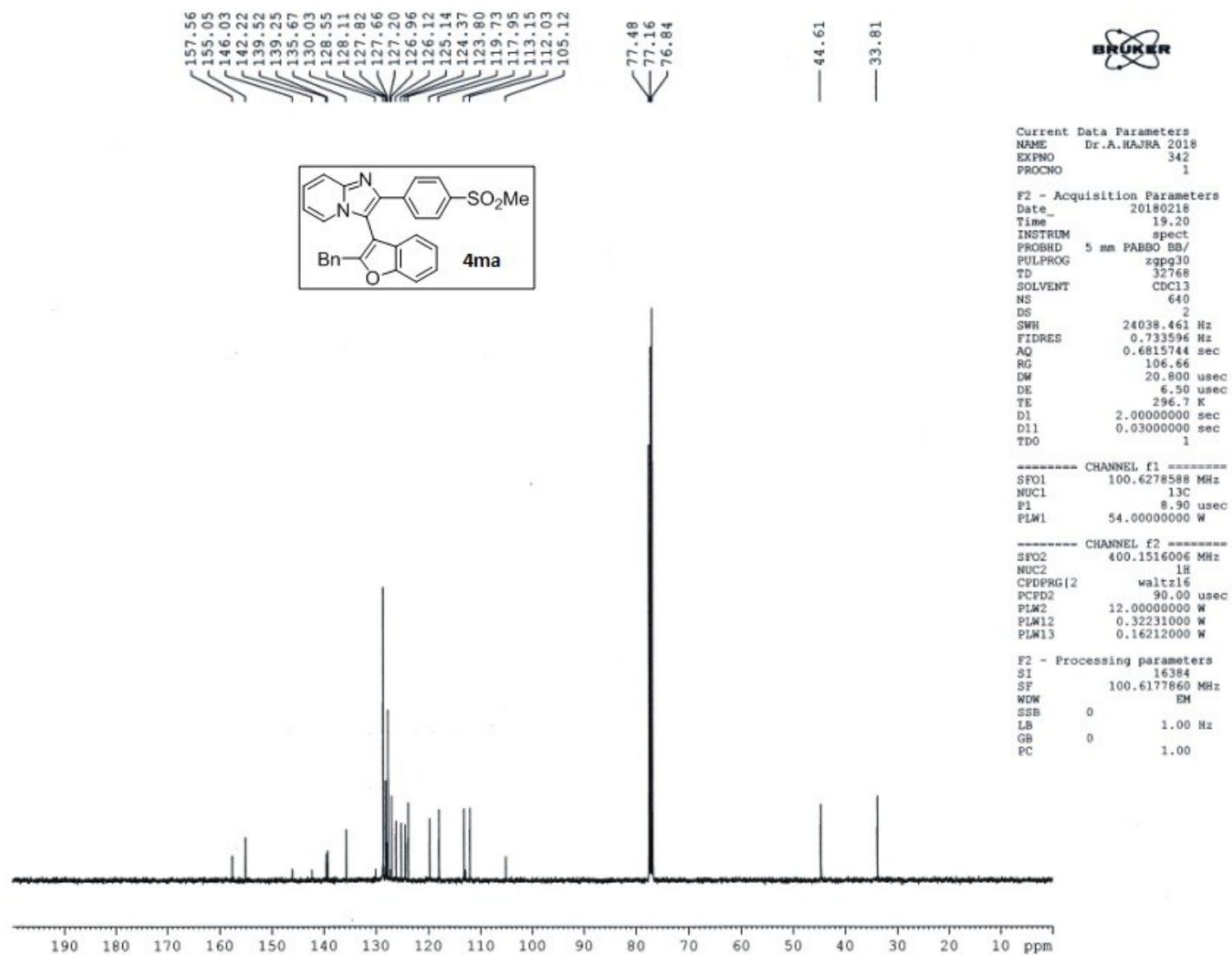
F2 - Acquisition Parameters
 Date_ 20170710
 Time_ 18.43
 INSTRUM spect
 PROBHD 5 mm PABBO BB/
 PULPROG zgpg30
 TD 32768
 SOLVENT CDCl3
 NS 250
 DS 2
 SWH 24038.461 Hz
 FIDRES 0.733596 Hz
 AQ 0.6815744 sec
 RG 77.59
 DW 20.800 usec
 DE 6.50 usec
 TE 298.6 K
 D1 2.00000000 sec
 D11 0.03000000 sec
 TD0 1

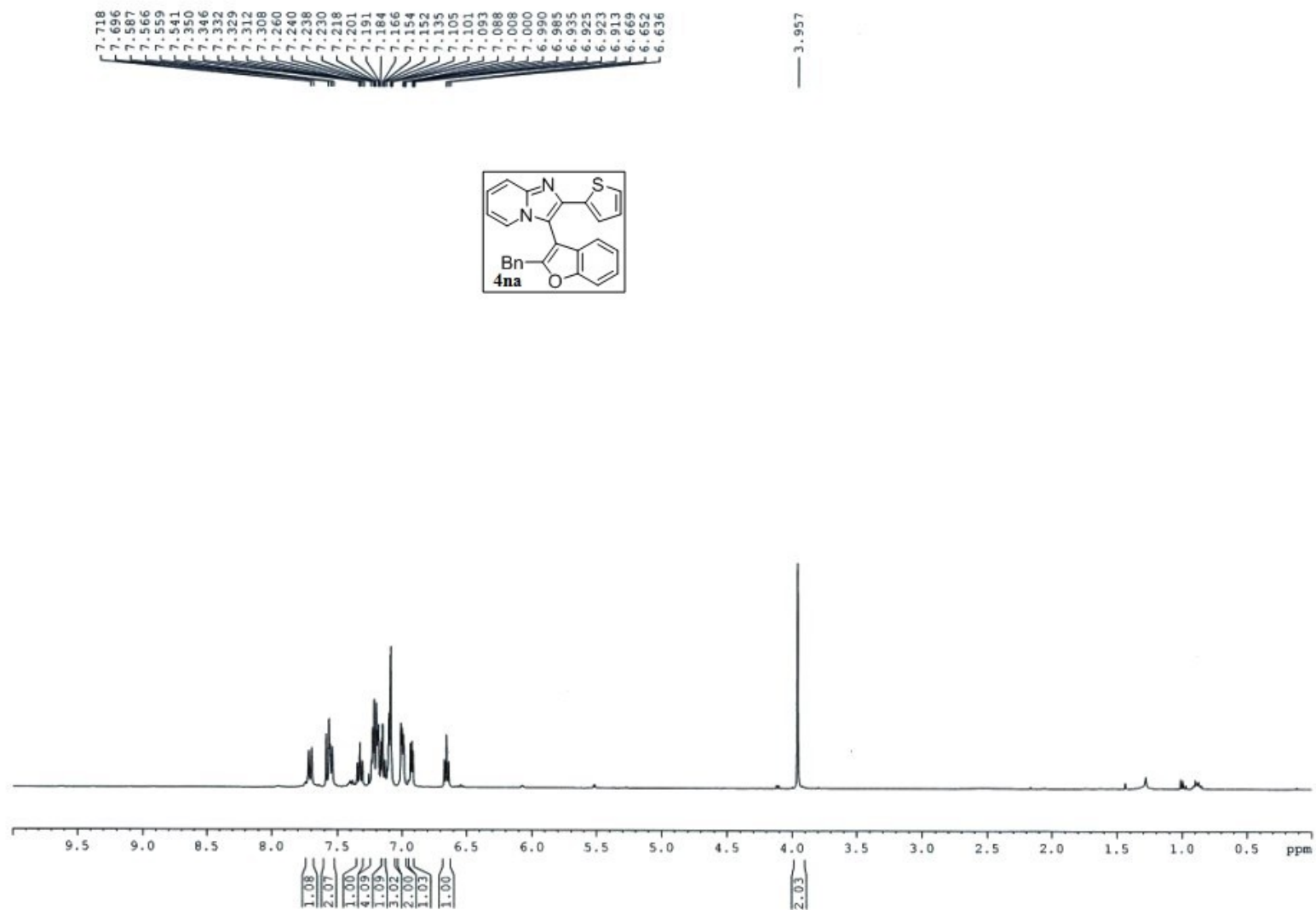
===== CHANNEL f1 =====
 SFO1 100.6278588 MHz
 NUC1 13C
 P1 8.90 usec
 PLW1 54.00000000 W

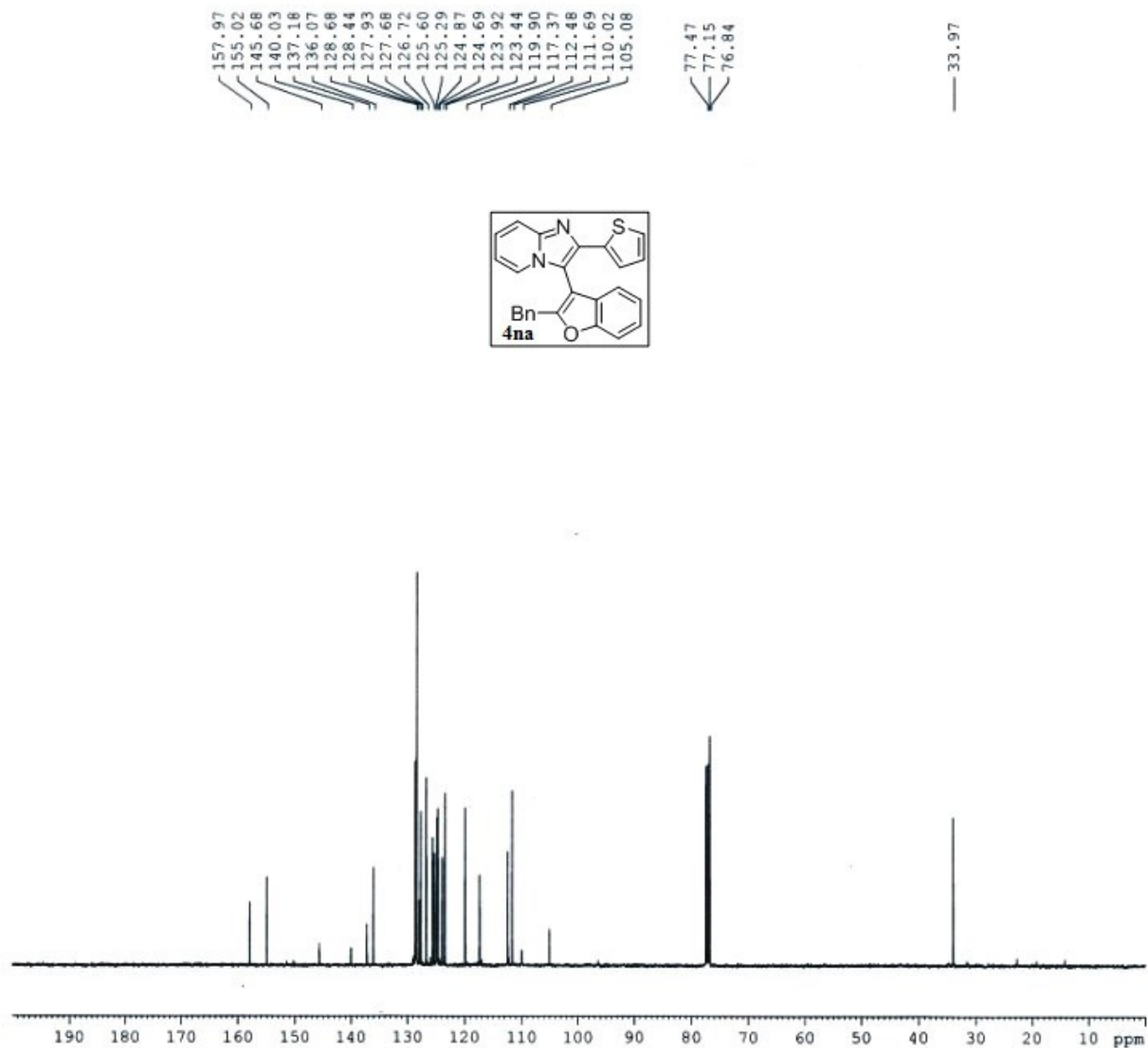
===== CHANNEL f2 =====
 SFO2 400.1516006 MHz
 NUC2 1H
 CPDPRG12 waltz16
 PCPD2 90.00 usec
 PLW2 12.00000000 W
 PLW12 0.32231000 W
 PLW13 0.16212000 W

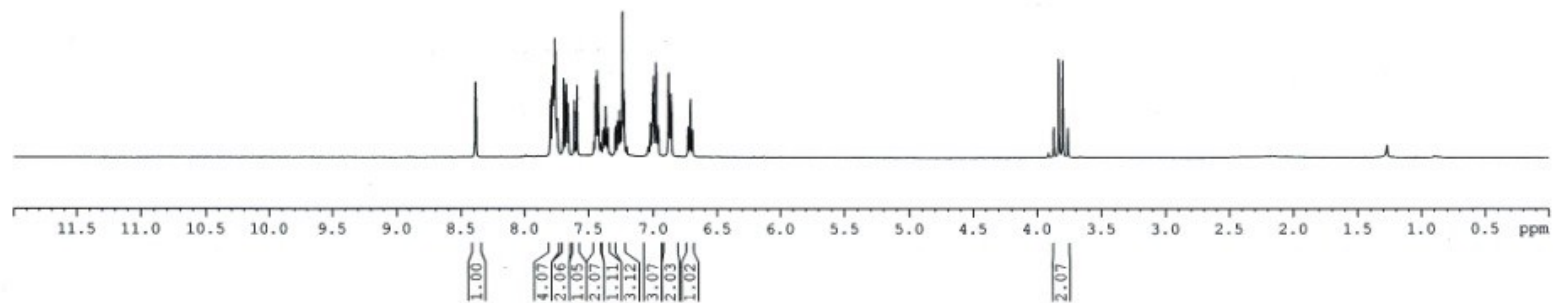
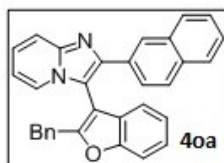
F2 - Processing parameters
 SI 16384
 SF 100.6177864 MHz
 WDW EM
 SSB 0
 LB 1.00 Hz
 GB 0
 FC 1.40

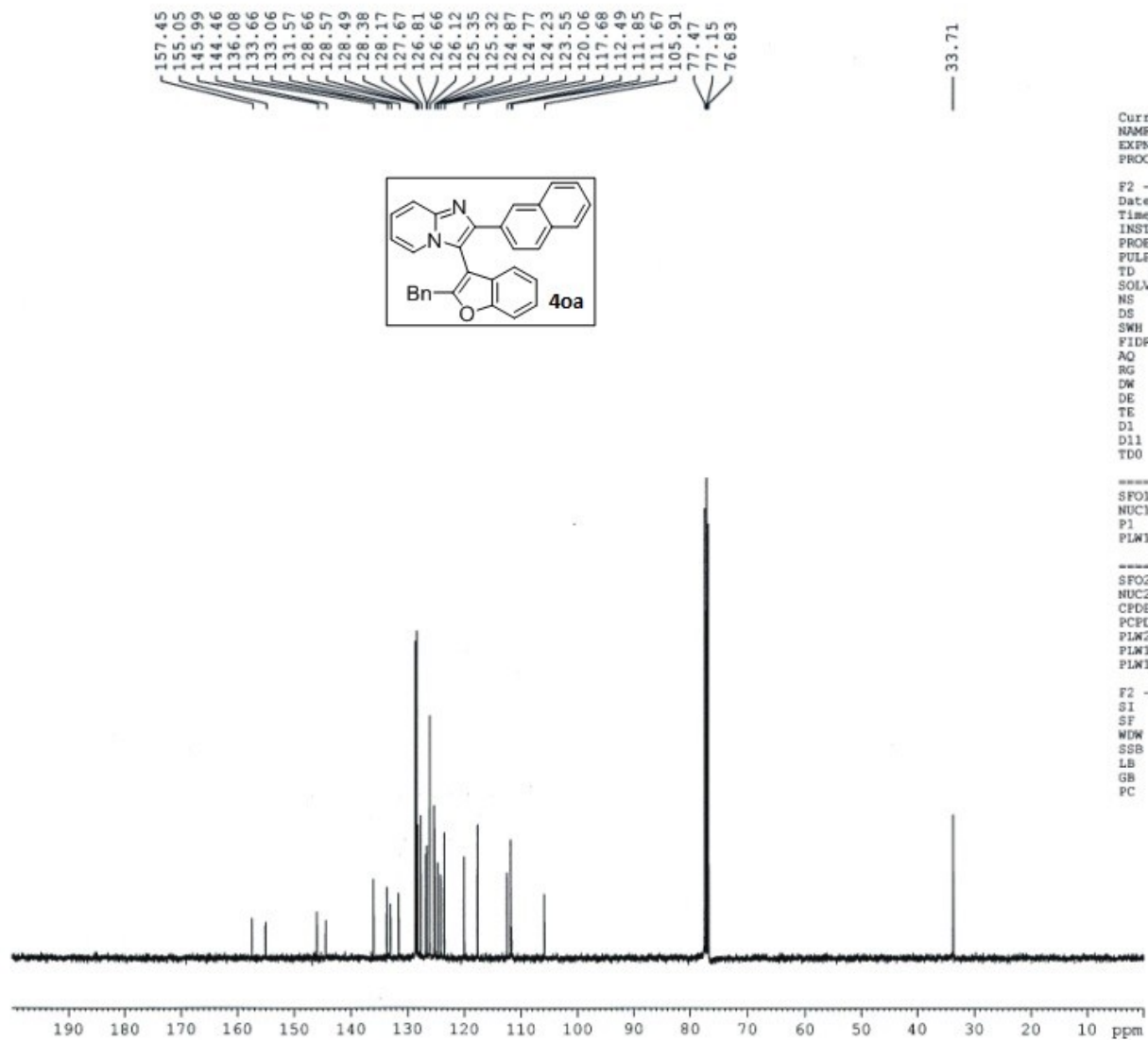












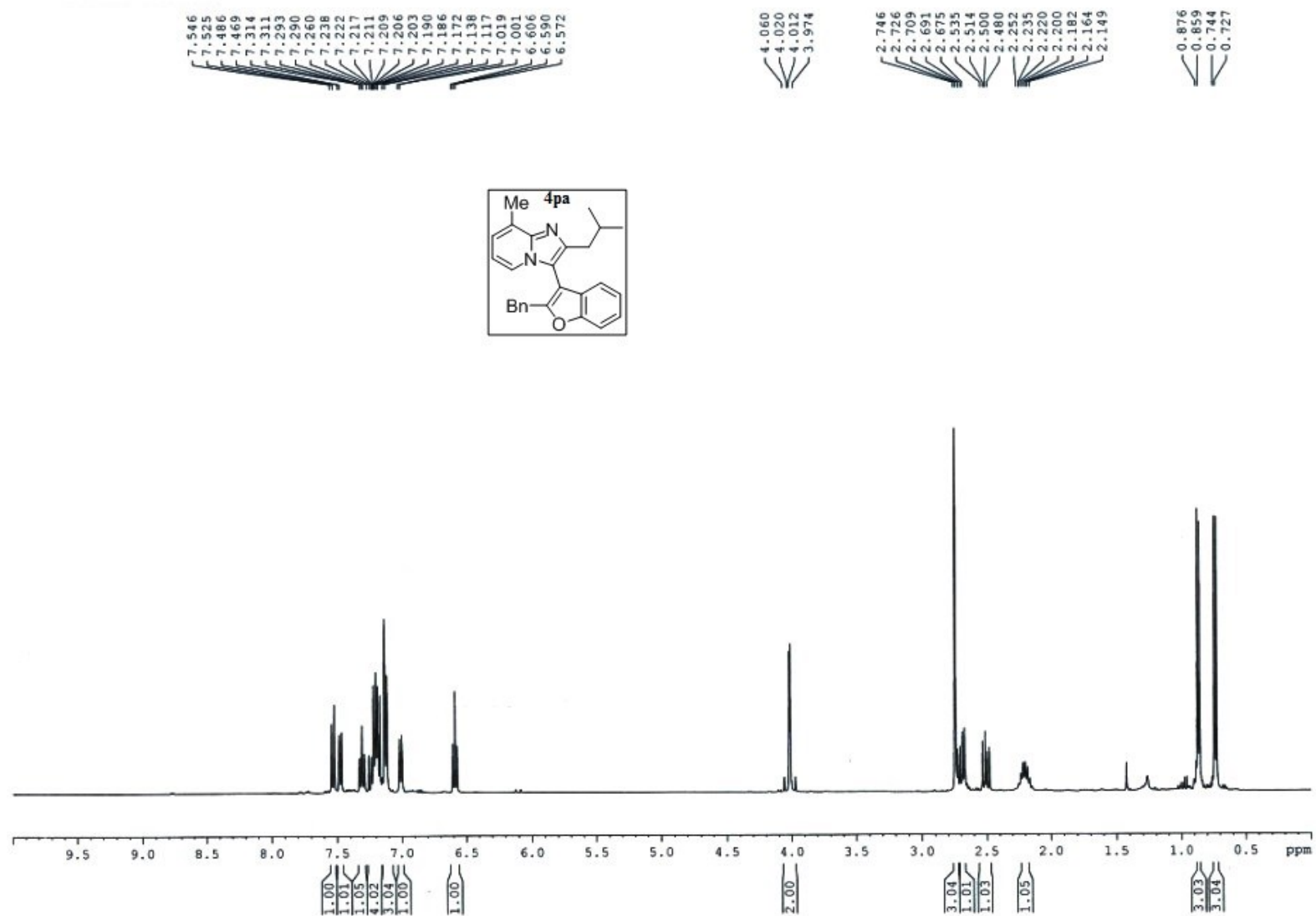
Current Data Parameters
 NAME Dr.A.HAJRA 2018
 EXPNO 341
 PROCNO 1

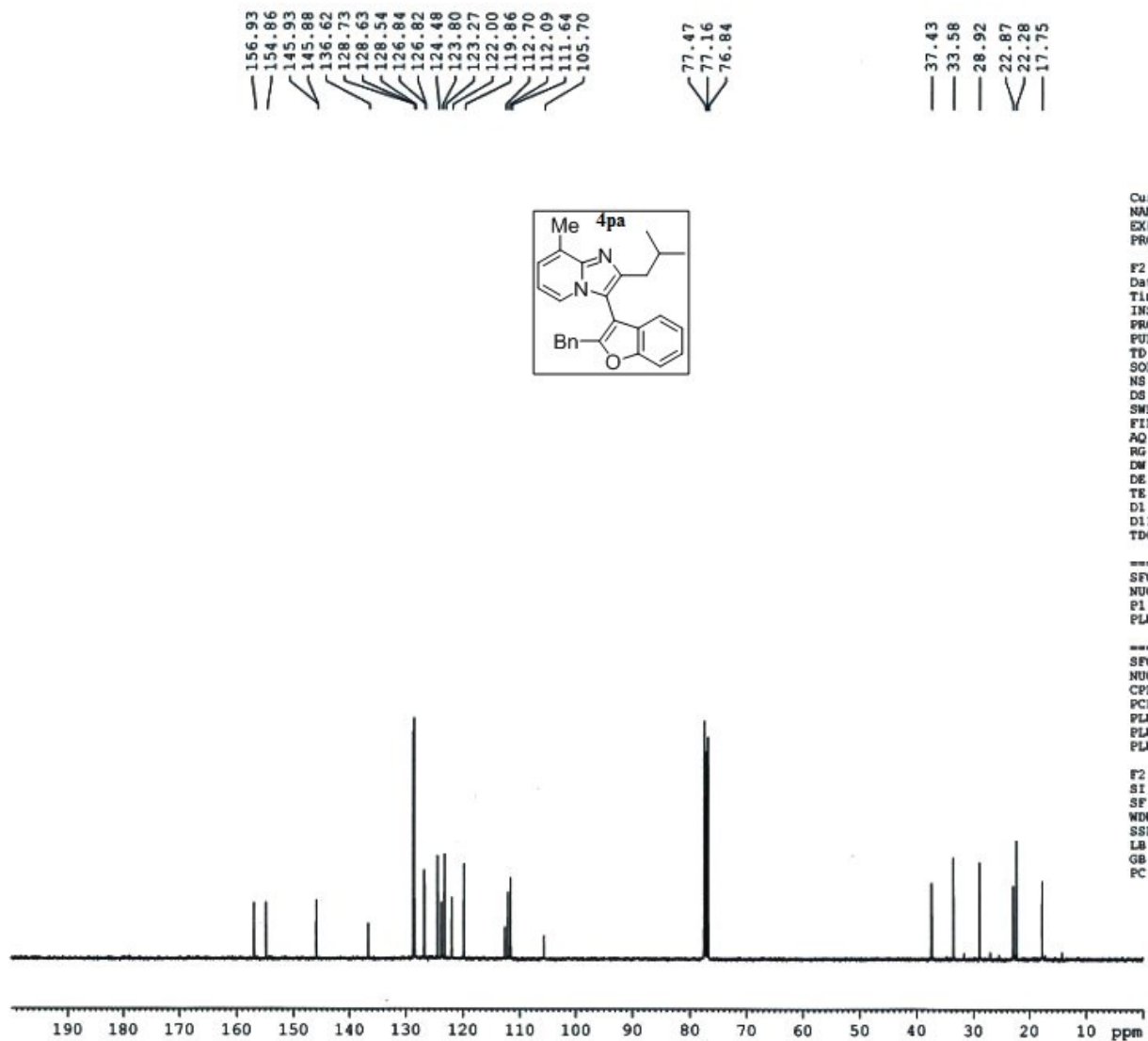
F2 - Acquisition Parameters
 Date 20180218
 Time 18.29
 INSTRUM spect
 PROBHD 5 mm PABBO BB/
 PULPROG zgpg30
 TD 32768
 SOLVENT CDCl3
 NS 330
 DS 2
 SWH 24038.461 Hz
 FIDRES 0.733596 Hz
 AQ 0.6815744 sec
 RG 106.66
 DW 20.800 usec
 DE 6.50 usec
 TE 296.5 K
 D1 2.00000000 sec
 D11 0.03000000 sec
 TD0 1

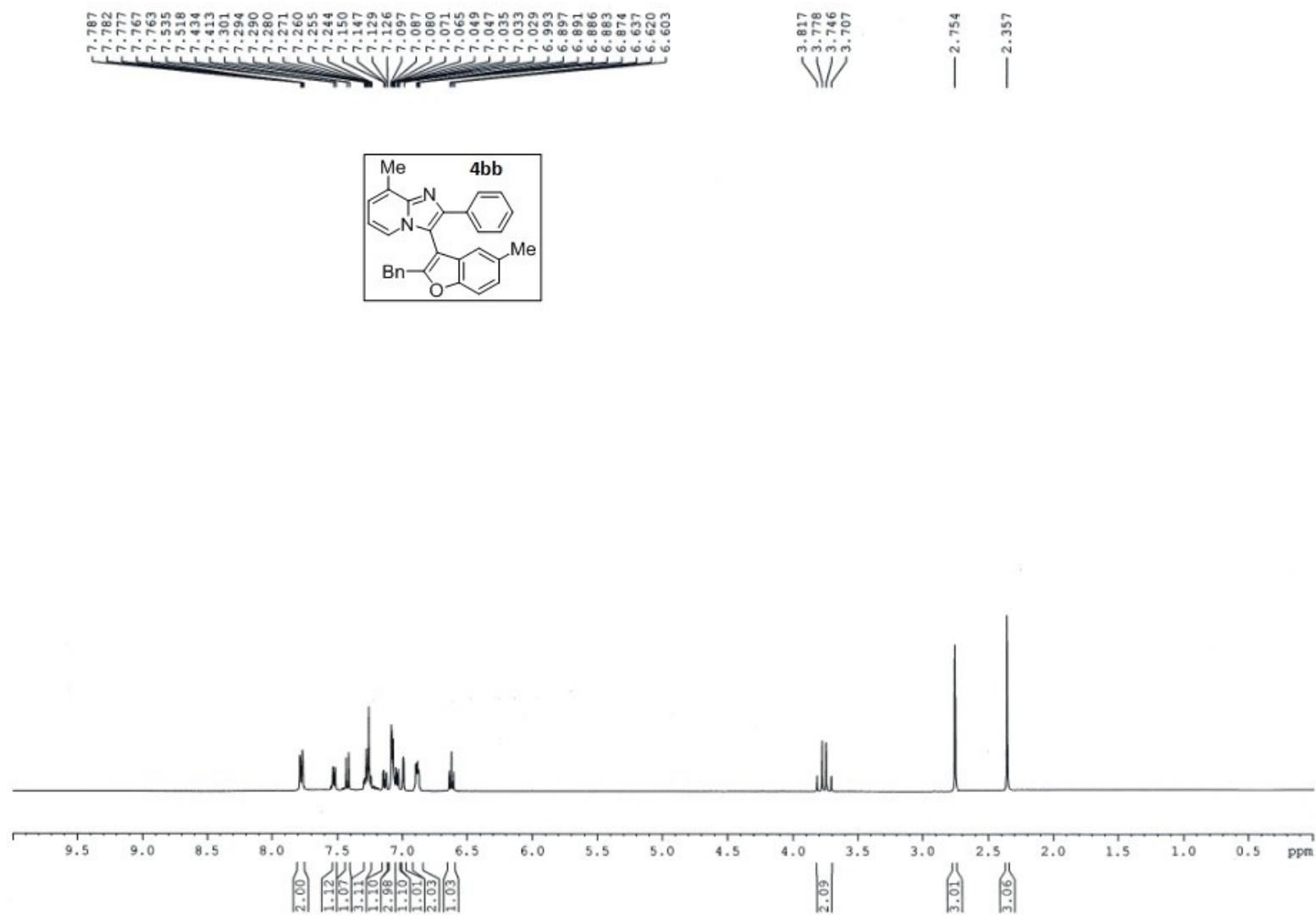
----- CHANNEL f1 -----
 SFO1 100.6278588 MHz
 NUC1 13C
 P1 8.90 usec
 PLW1 54.00000000 W

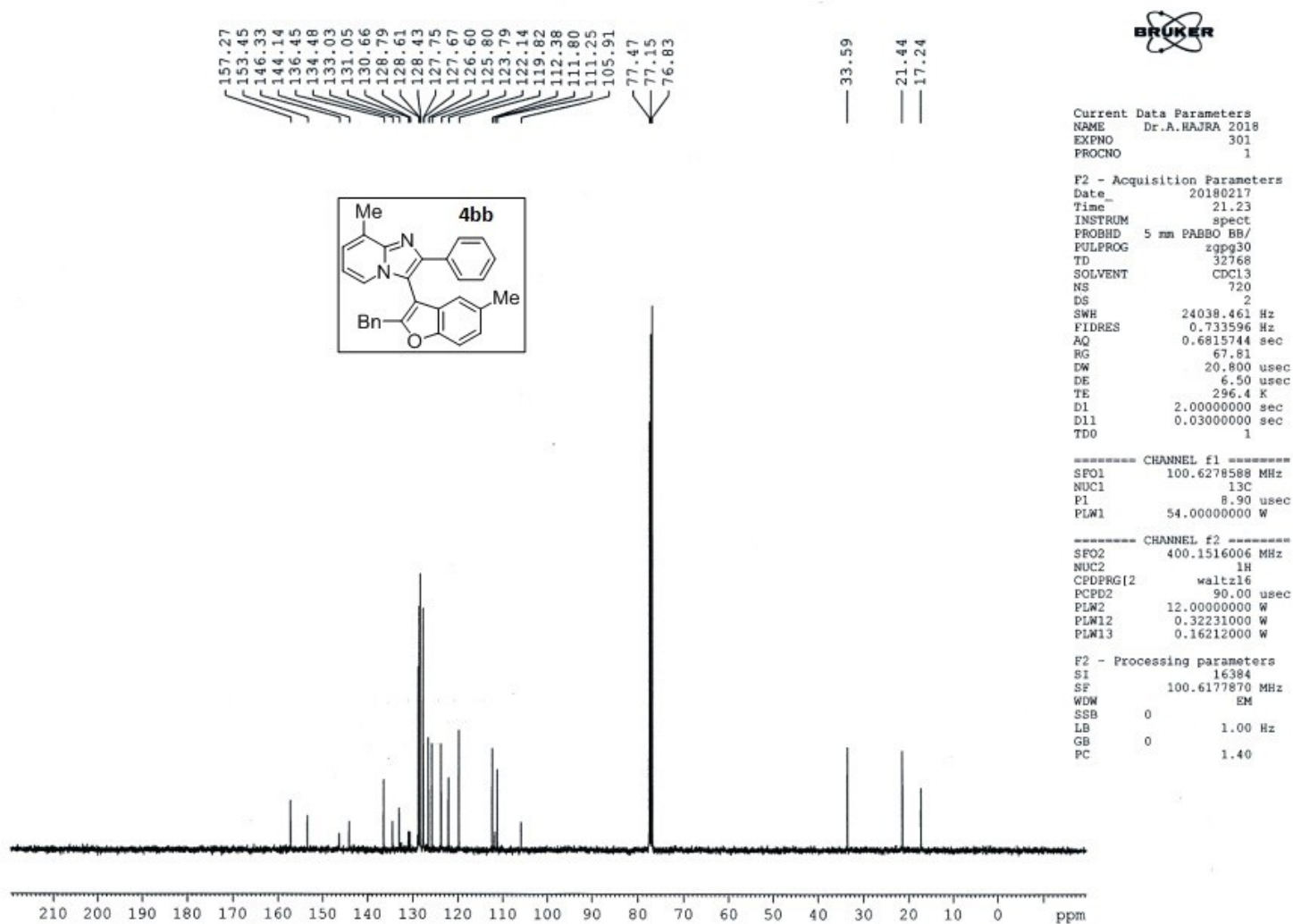
----- CHANNEL f2 -----
 SFO2 400.1516006 MHz
 NUC2 1H
 CPDPRG2 waltz16
 PCPD2 90.00 usec
 PLW2 12.00000000 W
 PLW12 0.32231000 W
 PLW13 0.16212000 W

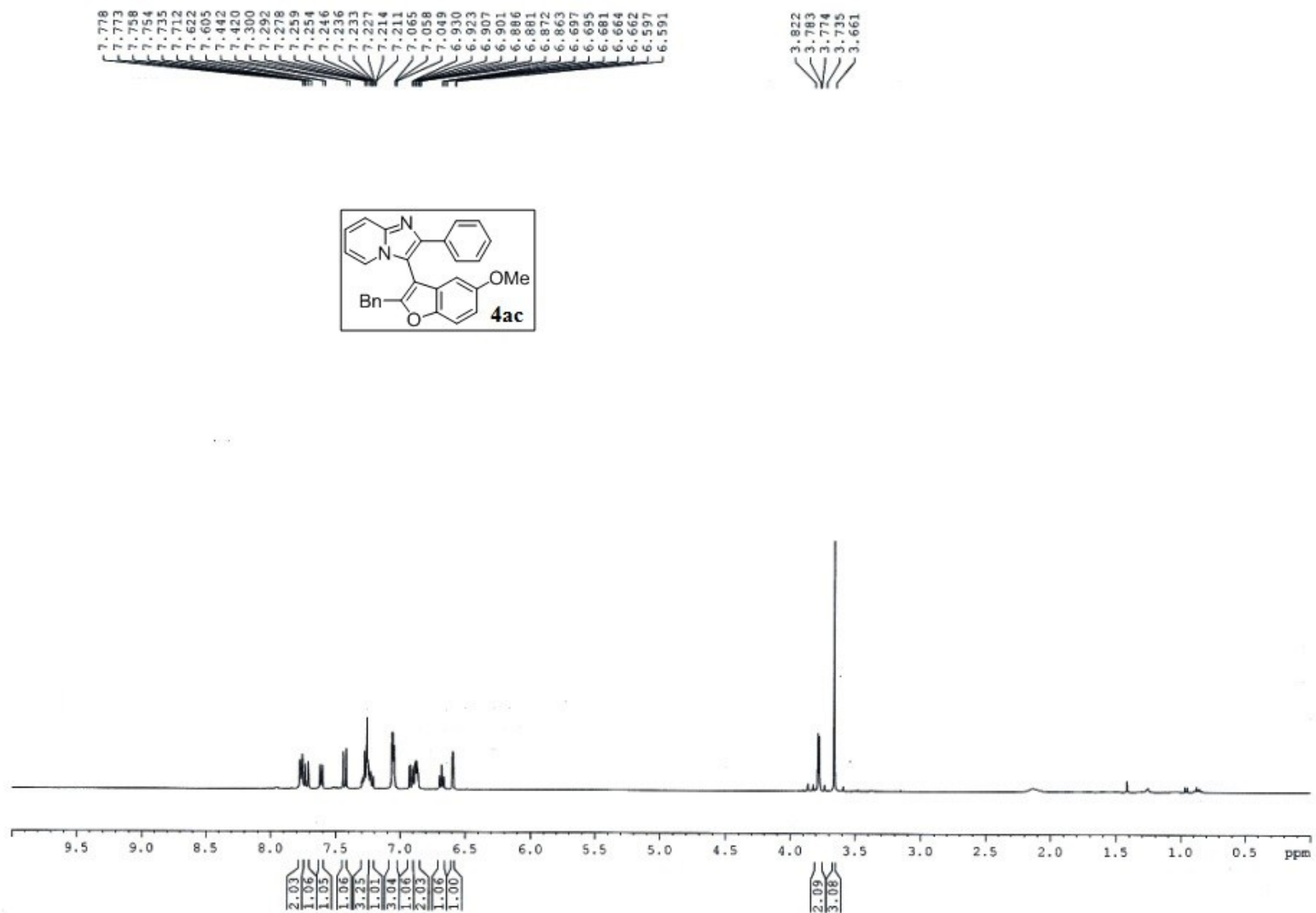
F2 - Processing parameters
 SI 16384
 SF 100.6177887 MHz
 WDW EM
 SSB 0
 LB 1.00 Hz
 GB 0
 PC 1.00

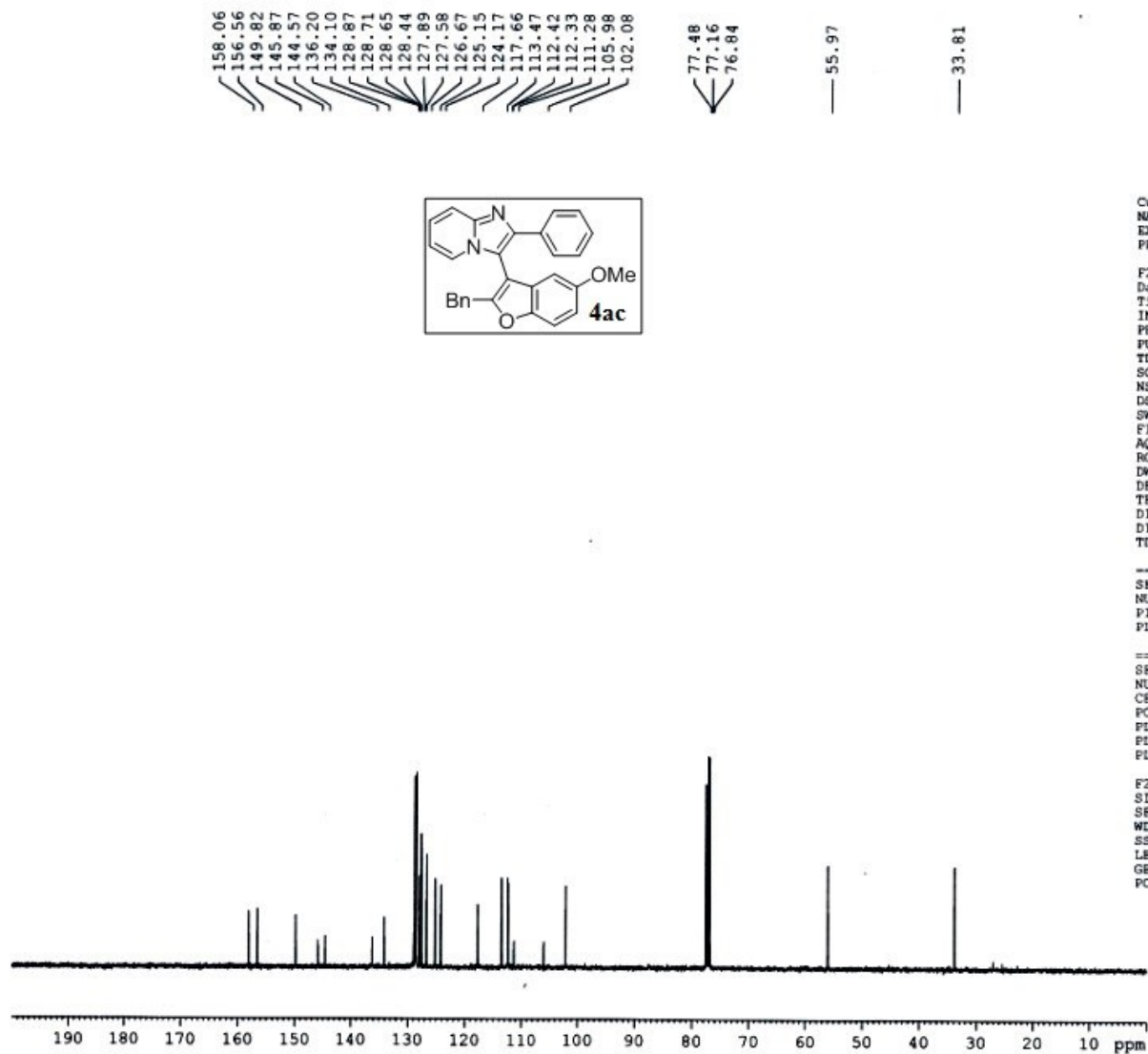












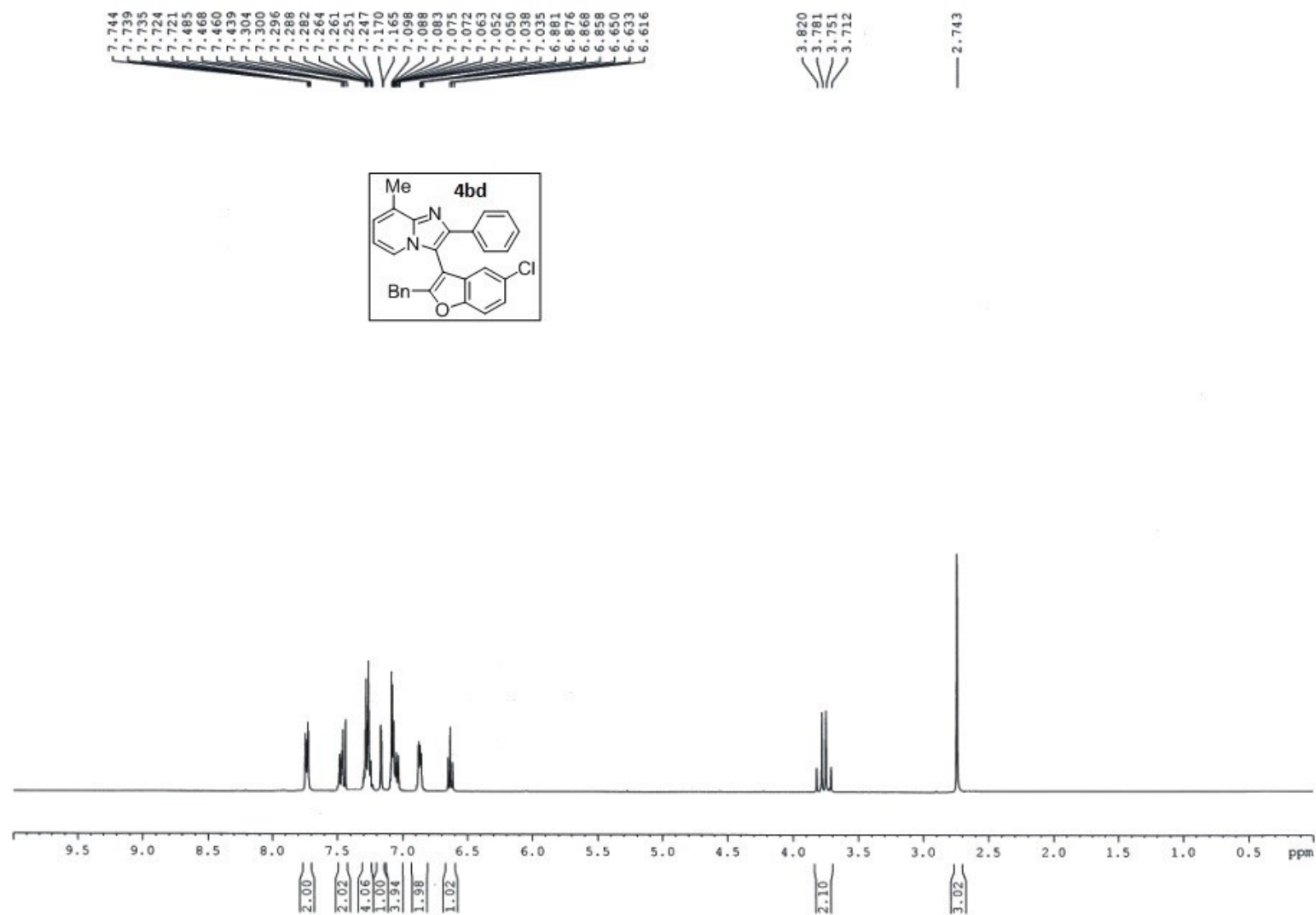
Current Data Parameters
NAME Dr.A.KAJRA 2017
EXPNO 1207
PROCNO 1

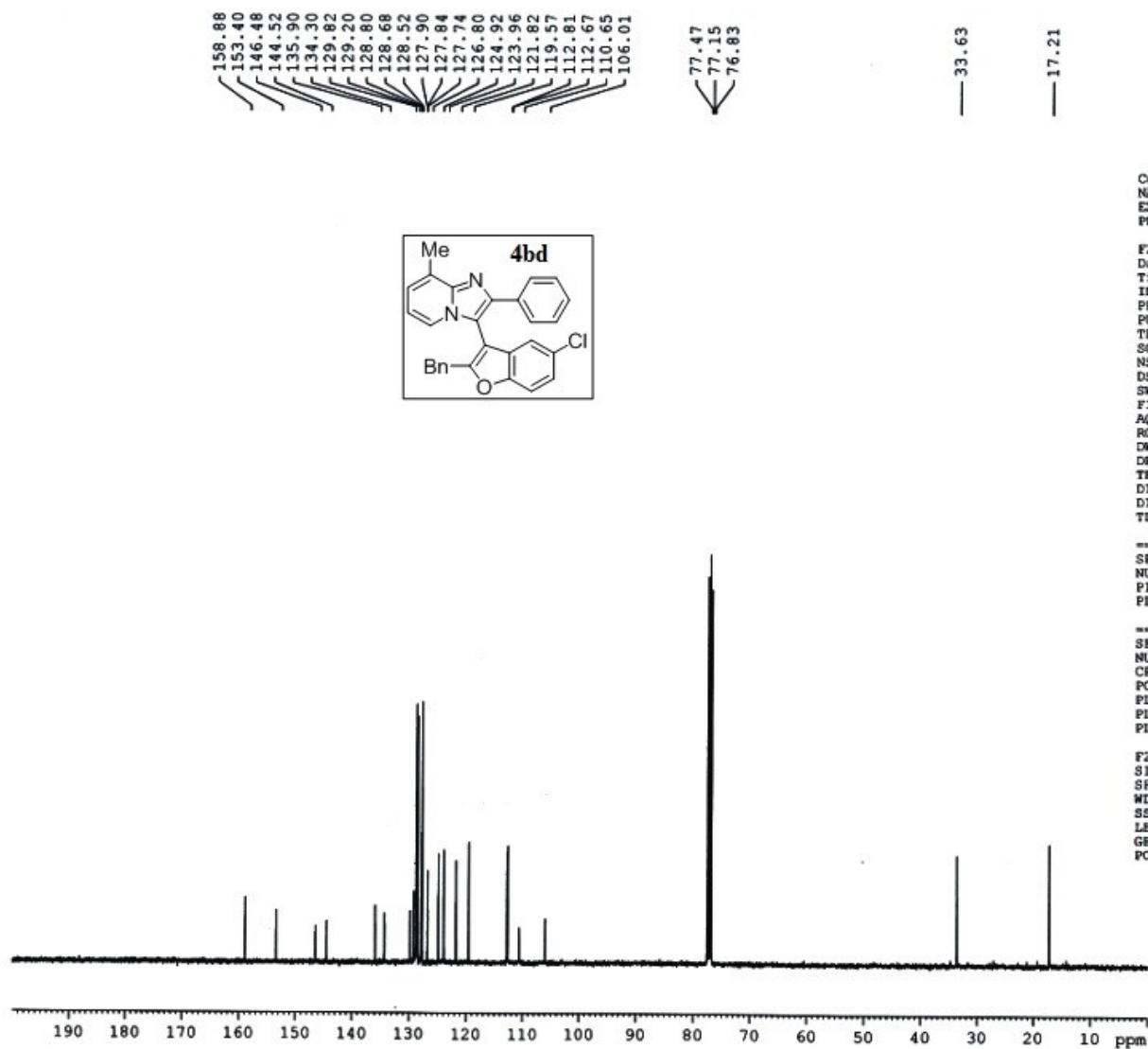
F2 - Acquisition Parameters
Date_ 20170715
Time 11.25
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDCl3
NS 200
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 77.59
DW 20.800 usec
DE 6.50 usec
TE 297.1 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

----- CHANNEL f1 -----
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

----- CHANNEL f2 -----
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG2 waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

F2 - Processing parameters
SI 16384
SF 100.6177895 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40





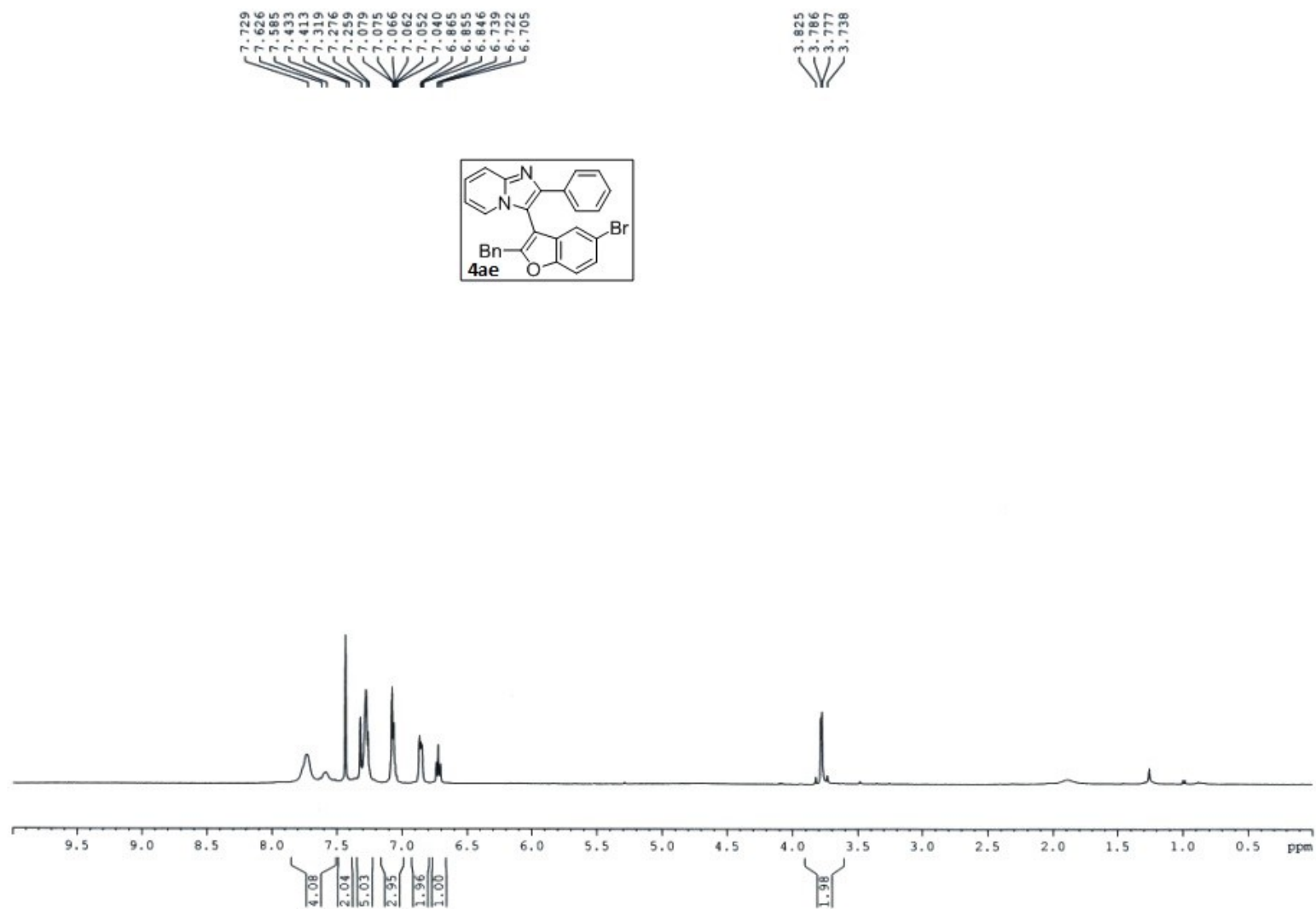
Current Data Parameters
NAME Dr.A.HAJRA 2016
EXPNO 1404
PROCNO 1

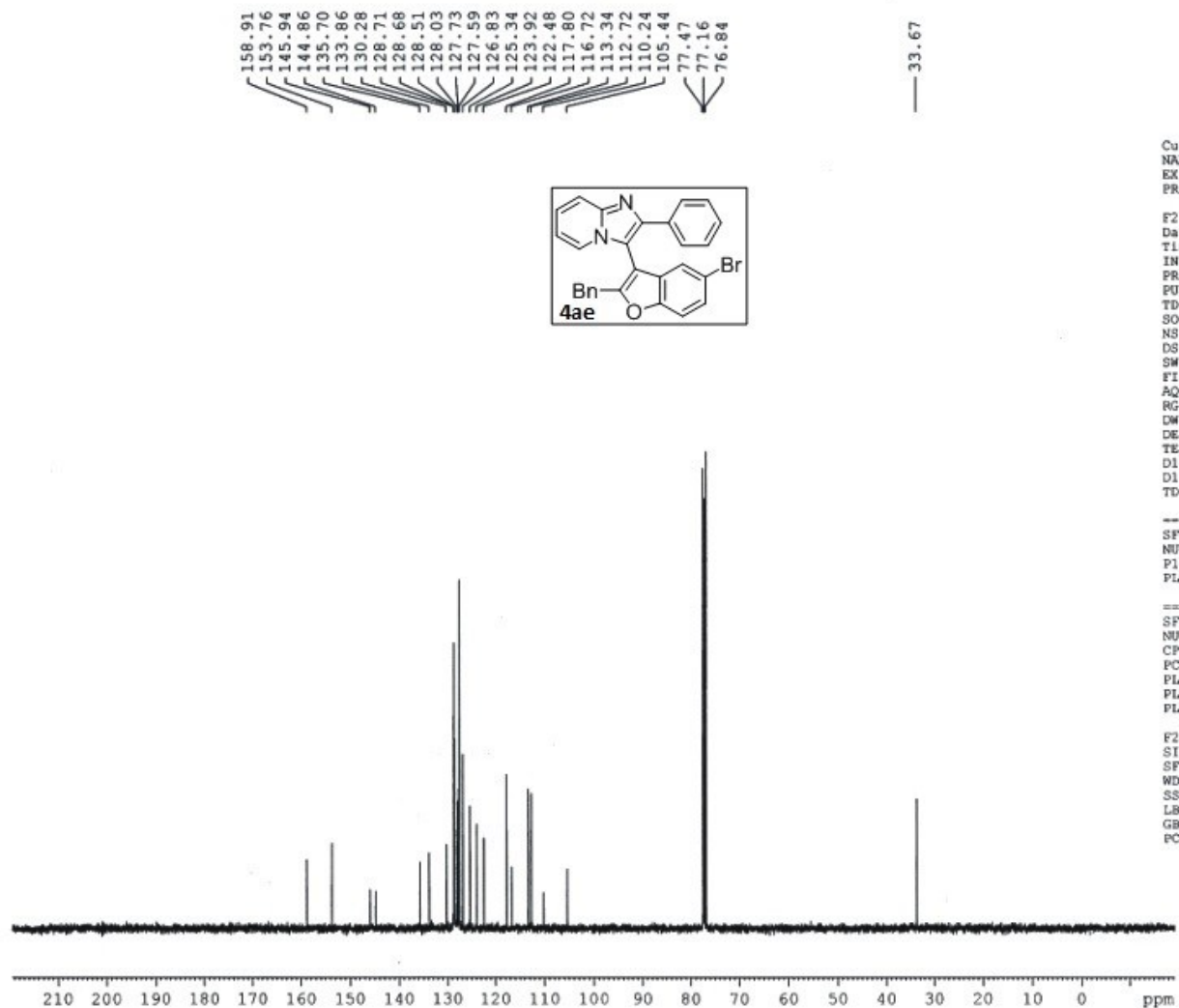
F2 - Acquisition Parameters
Date_ 20161218
Time_ 17.29
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDC13
NS 640
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 62.69
CW 20.800 usec
DE 6.50 usec
TE 298.0 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

===== CHANNEL f1 =====
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

===== CHANNEL f2 =====
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG2 waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

F2 - Processing parameters
SI 16384
SF 100.6177873 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40





— 33.67



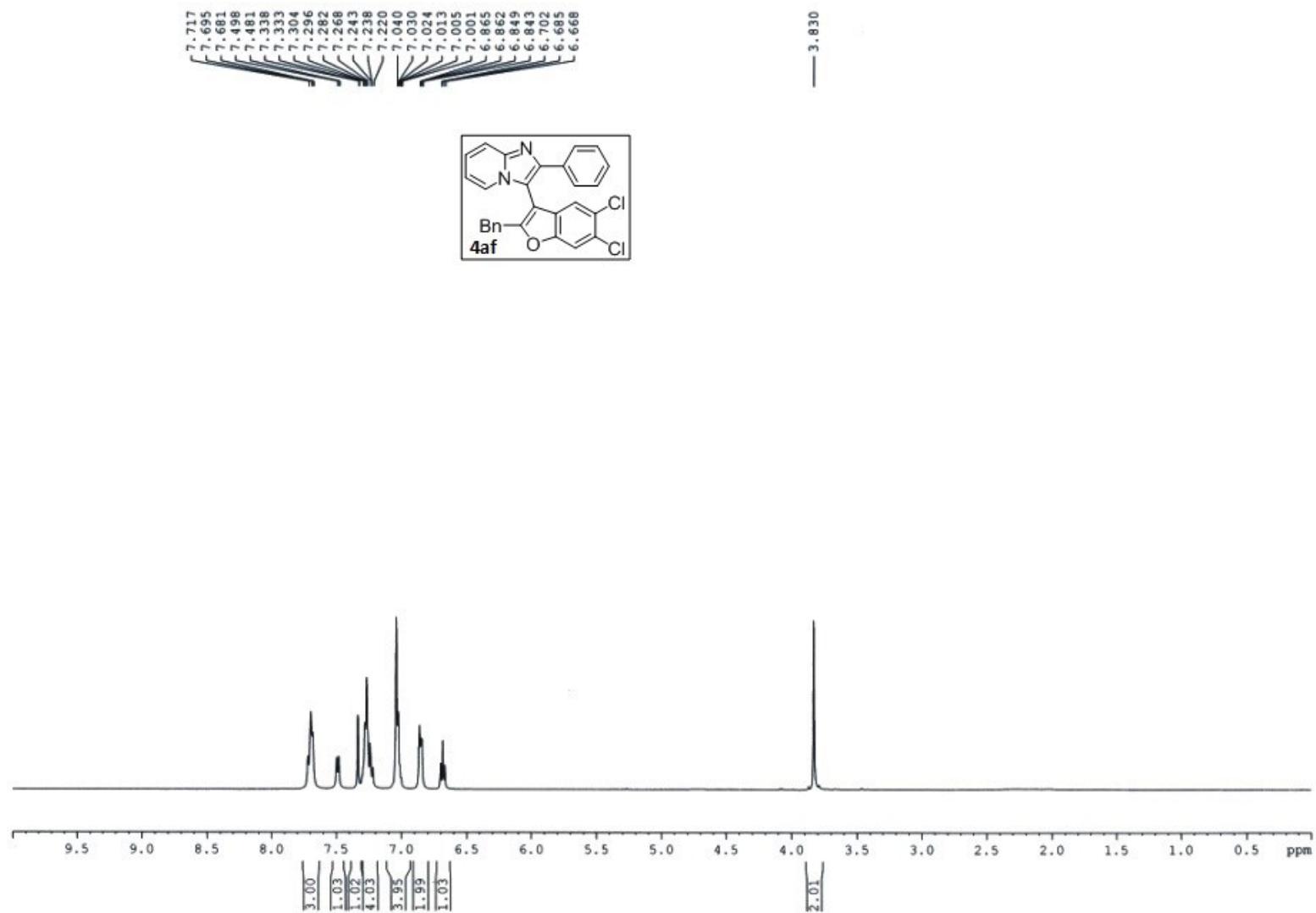
Current Data Parameters
NAME Dr.A.HAJRA 2018
EXPNO 304
PROCNO 1

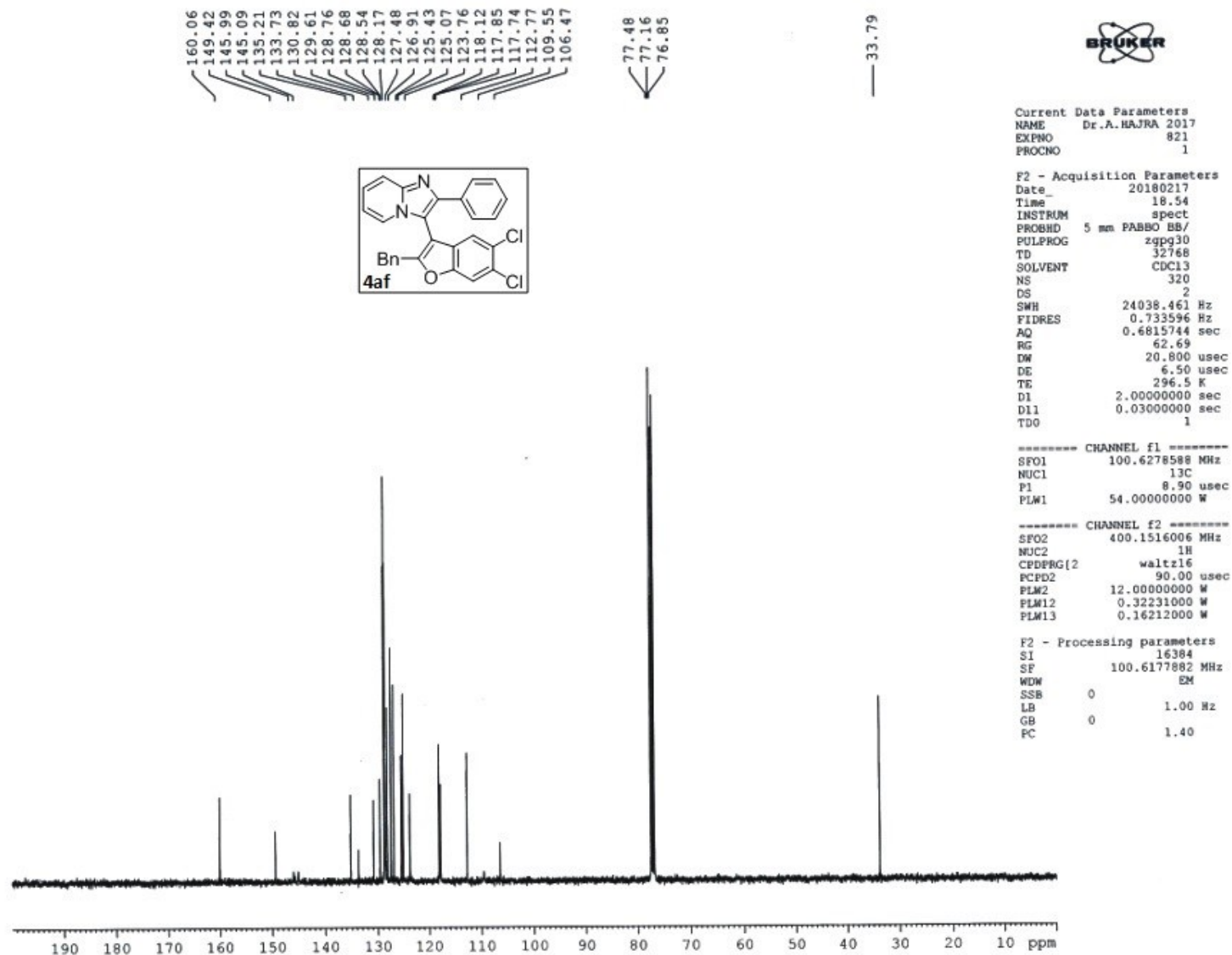
F2 - Acquisition Parameters
Date_ 20180218
Time 10.05
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDCl3
NS 240
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 106.66
DW 20.800 usec
DE 6.50 usec
TE 297.9 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

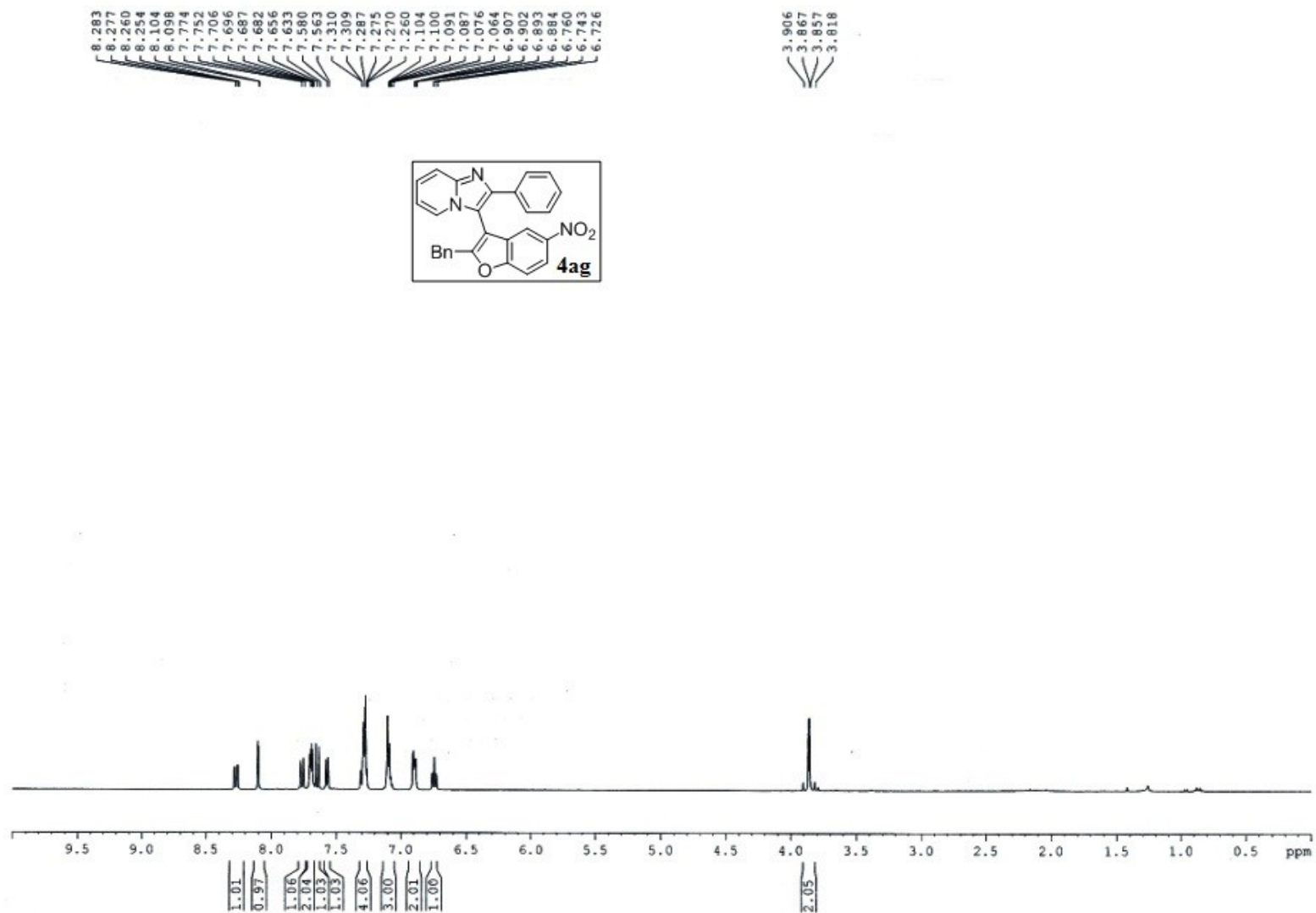
===== CHANNEL f1 =====
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

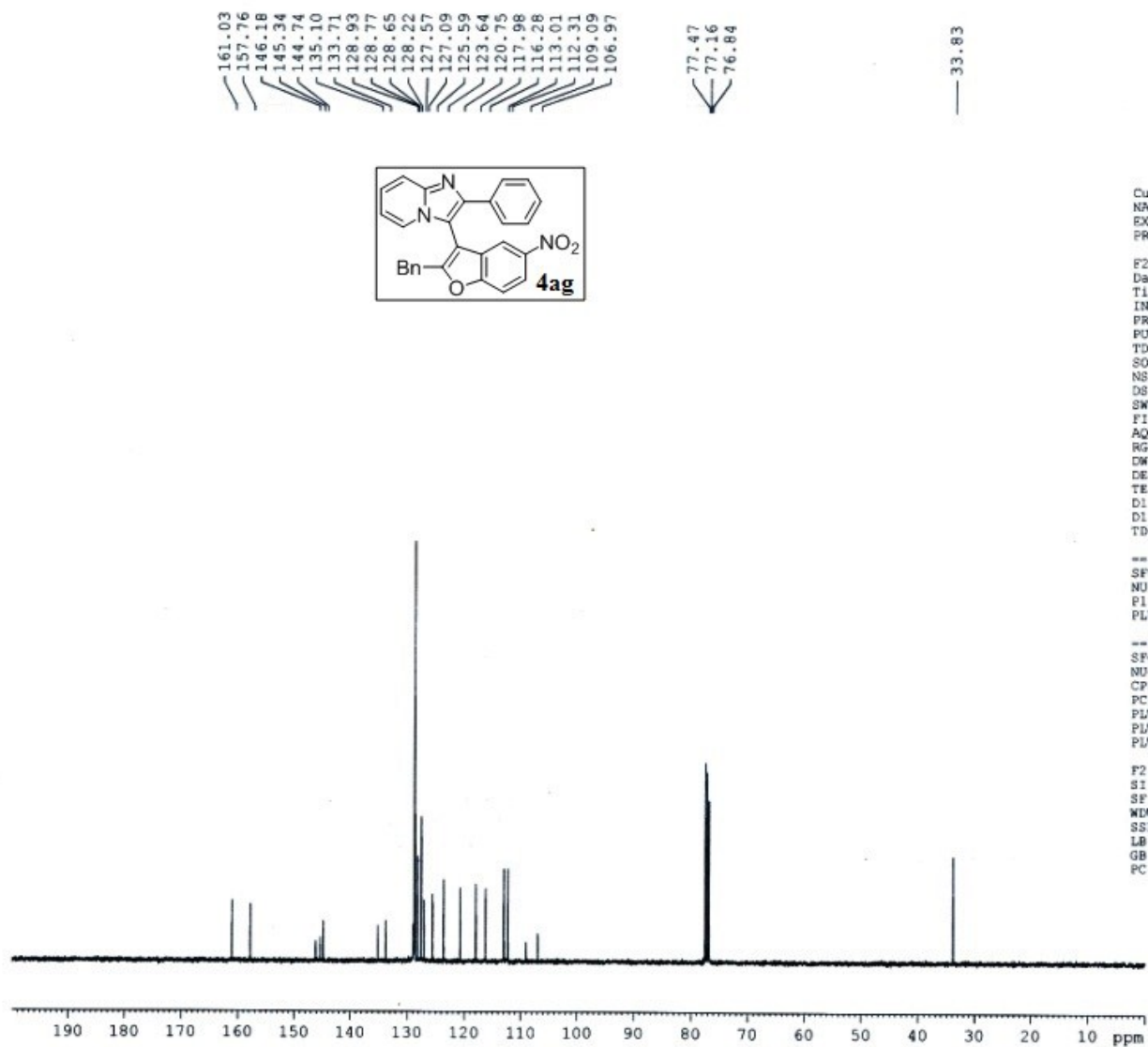
===== CHANNEL f2 =====
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG2 waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

F2 - Processing parameters
SI 16384
SF 100.6177891 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40









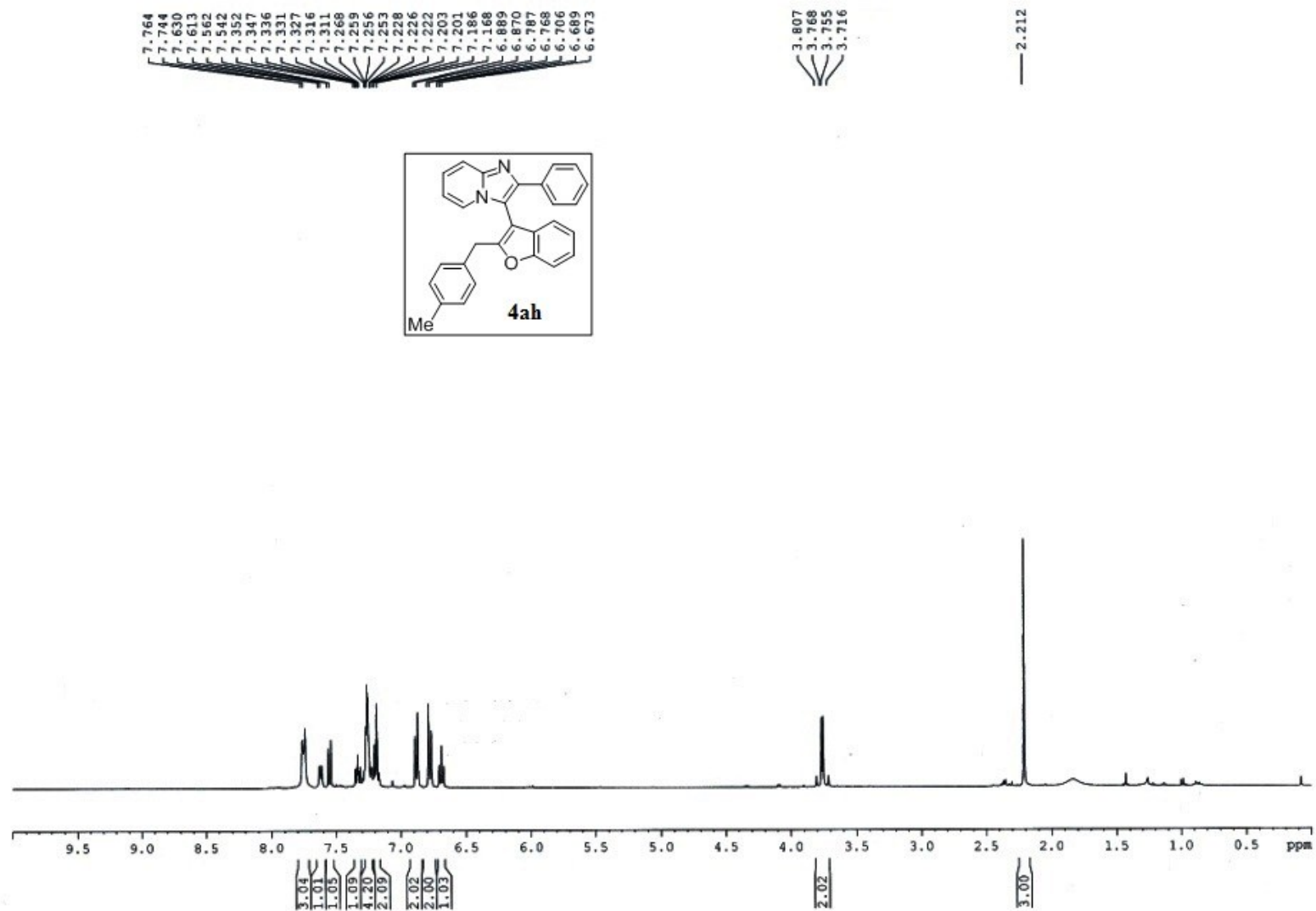
Current Data Parameters
NAME Dr.A.RAJRA 2017
EXPNO 542
PROCNO 1

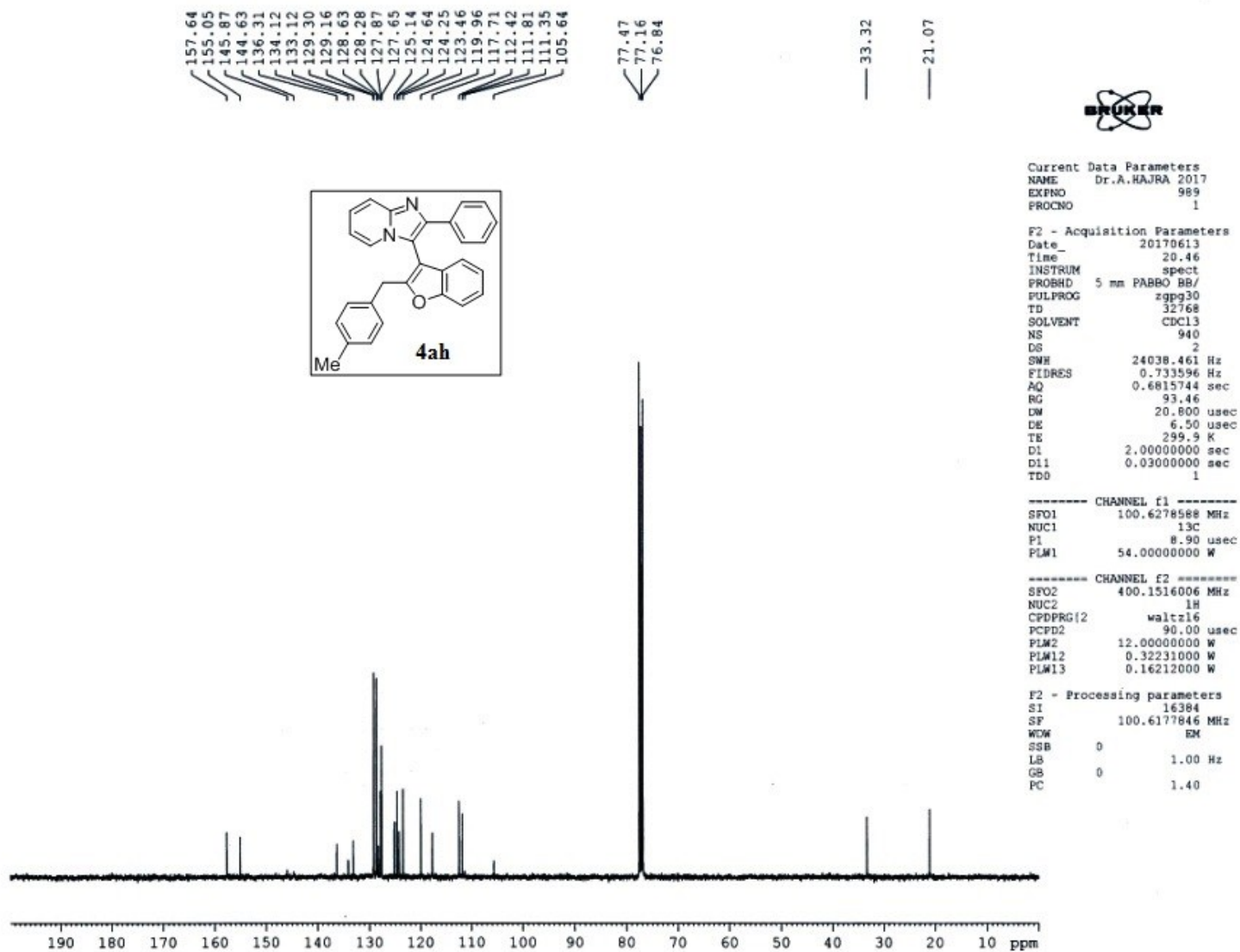
F2 - Acquisition Parameters
Date_ 20170718
Time 20.05
INSTRUM spect
PROBHD 5 mm PARBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDCl3
NS 200
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 62.69
CW 20.800 usec
DE 6.50 usec
TE 299.4 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

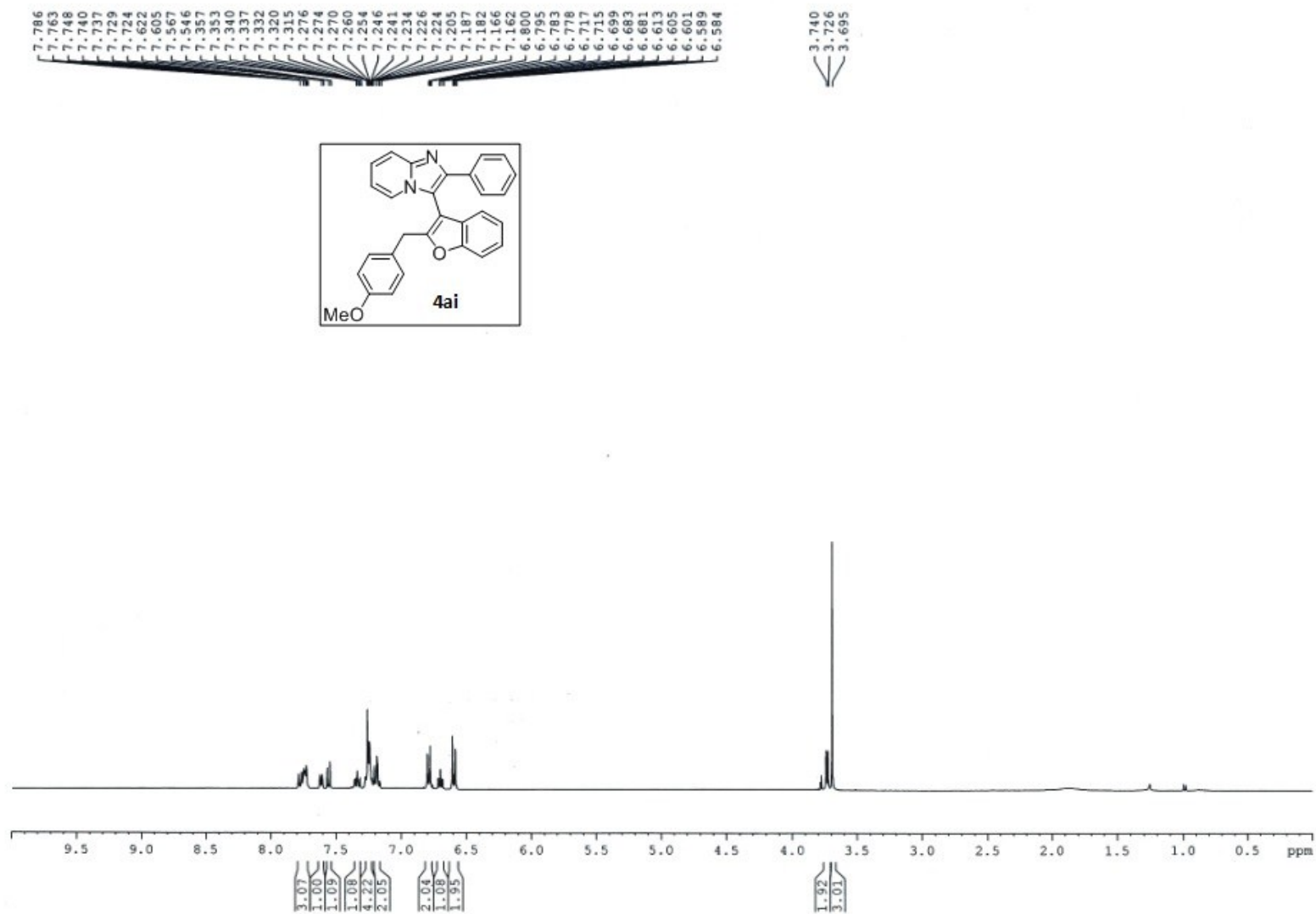
----- CHANNEL f1 -----
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

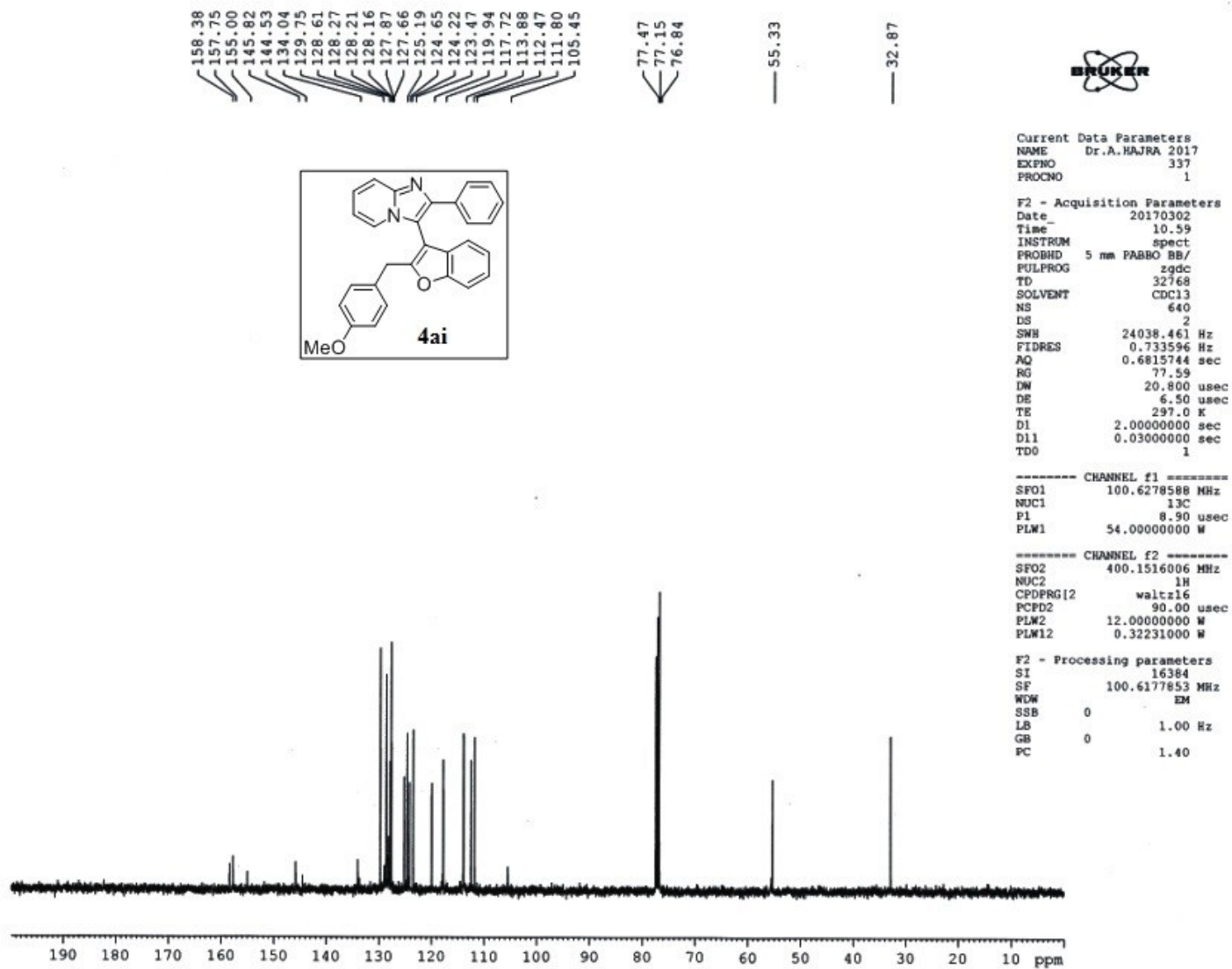
----- CHANNEL f2 -----
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG12 waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

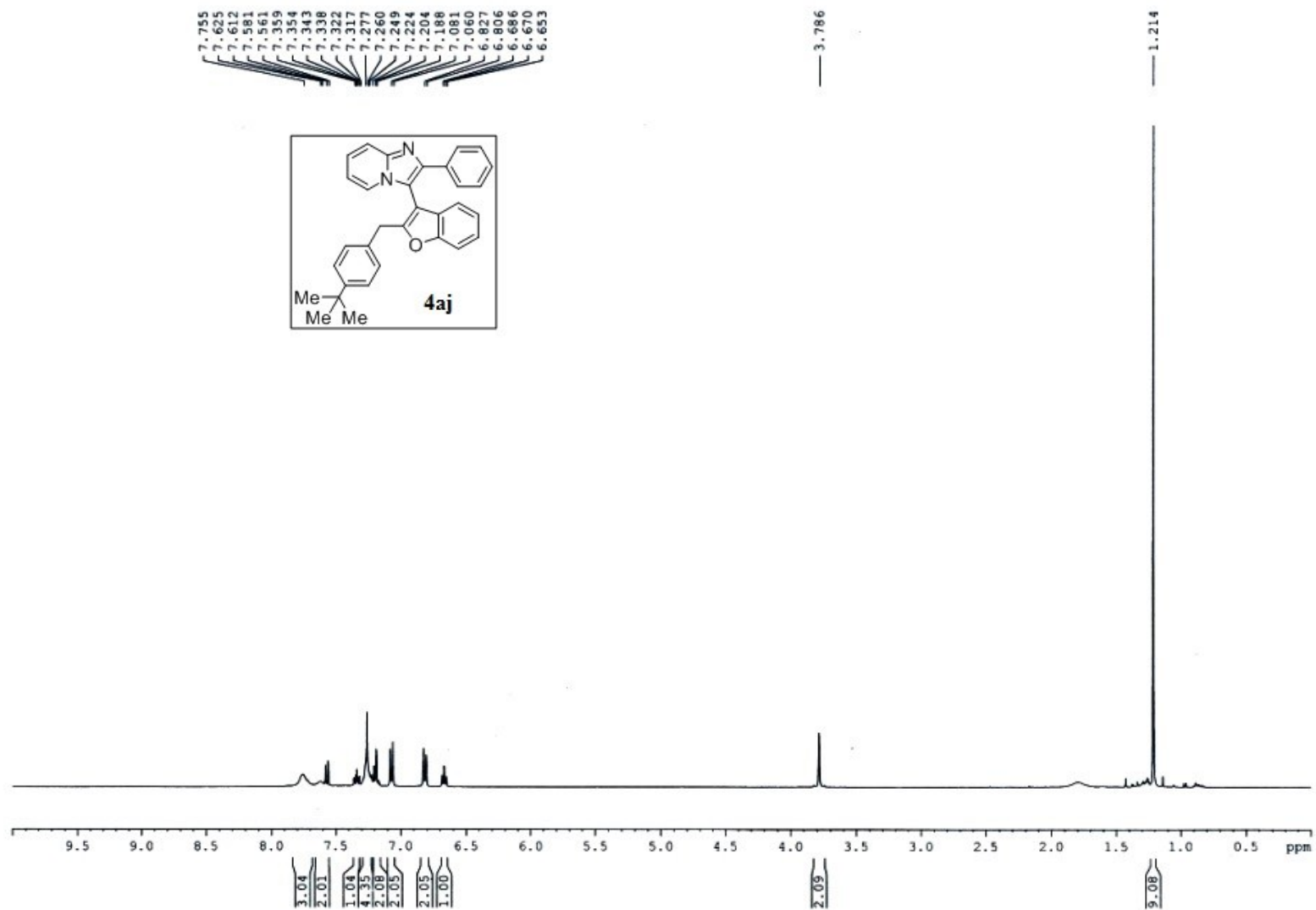
F2 - Processing parameters
SI 16384
SF 100.6177887 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

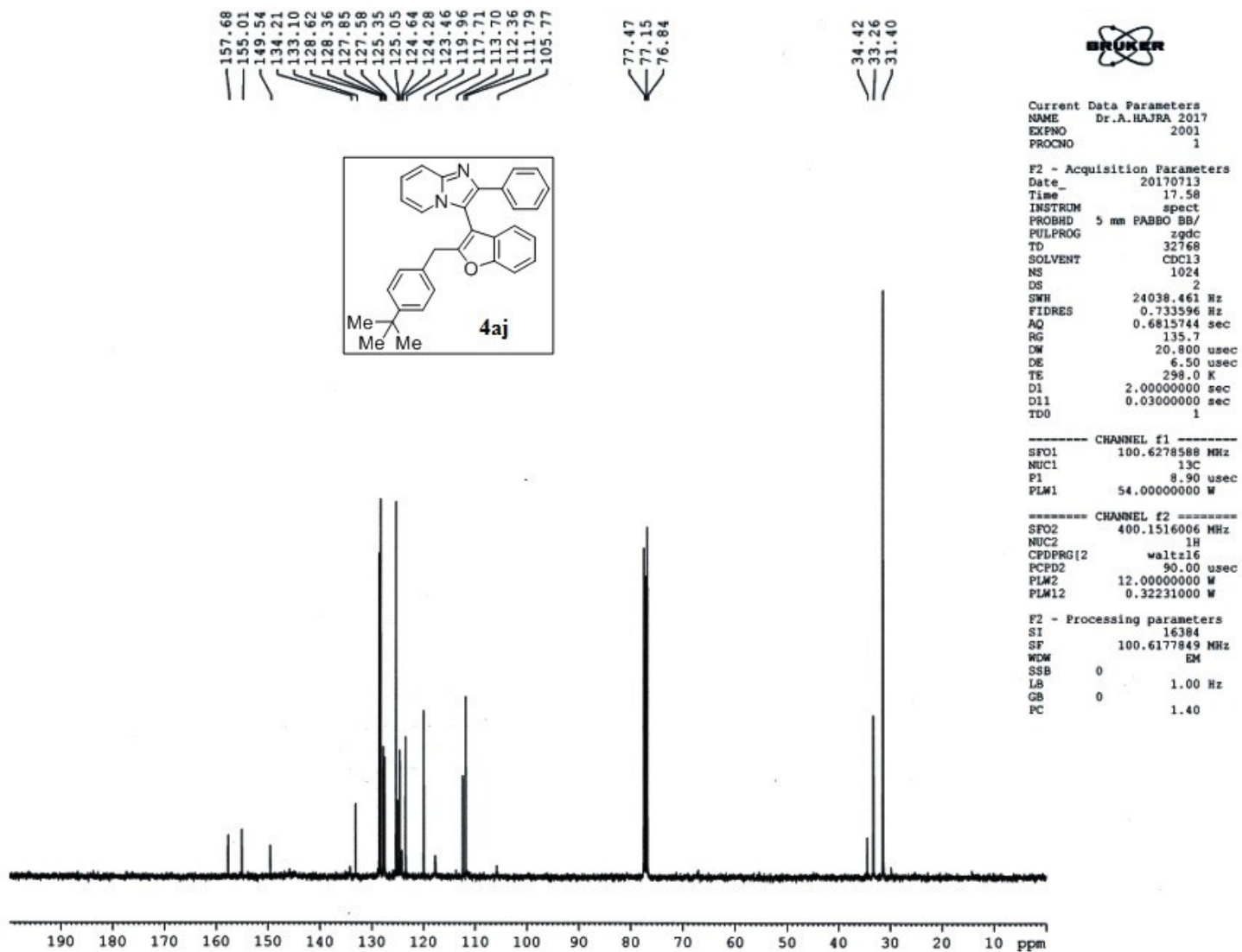


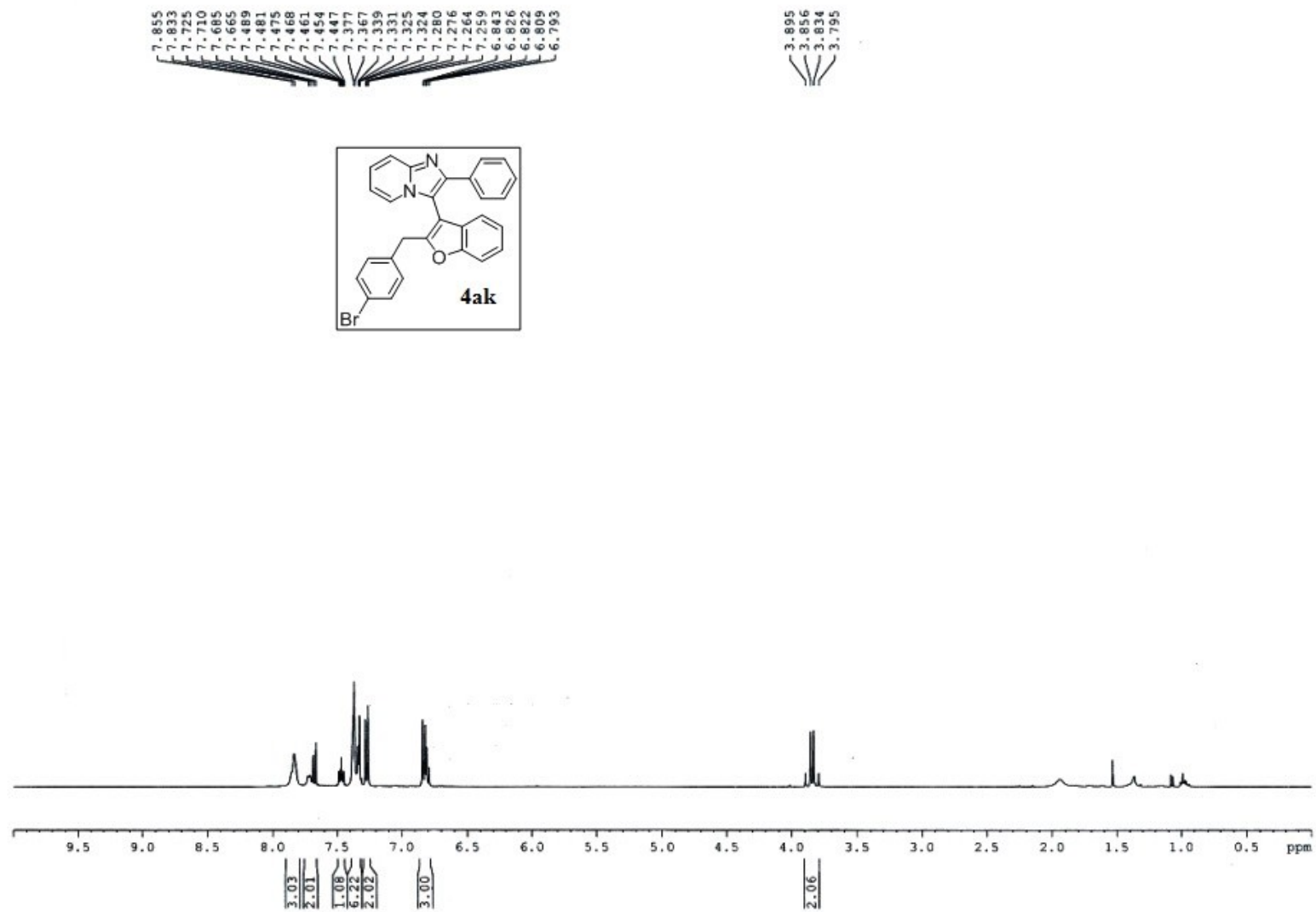


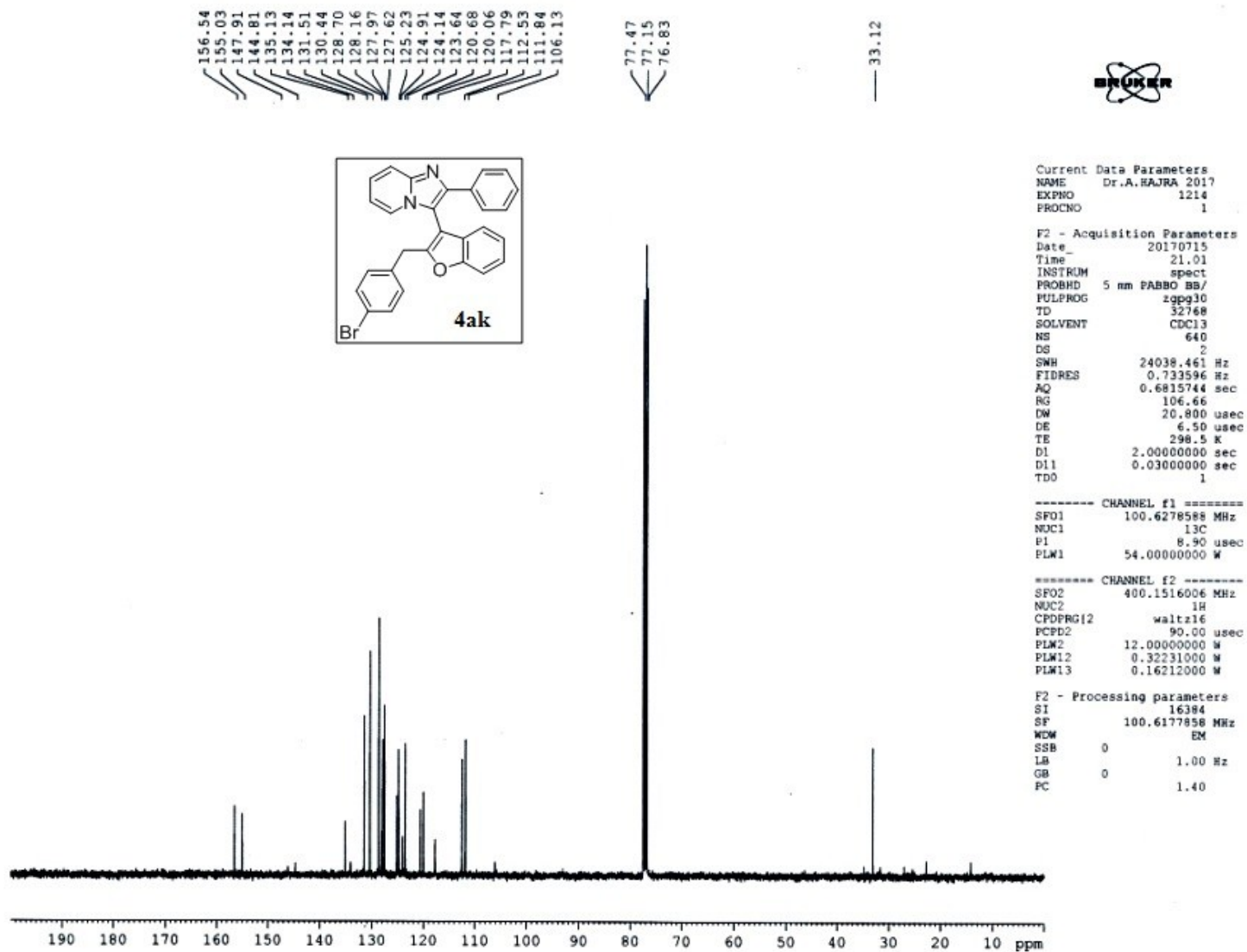


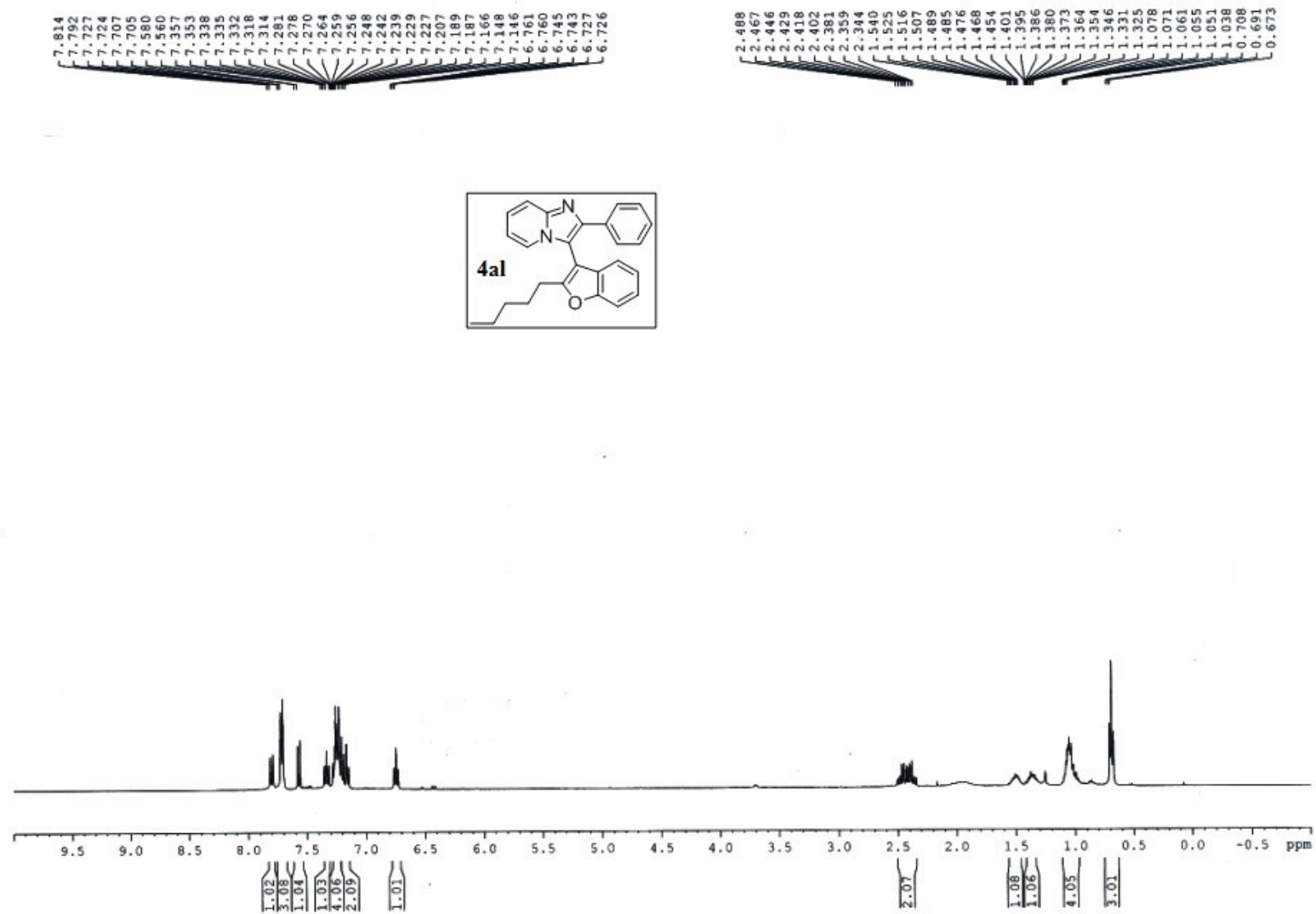


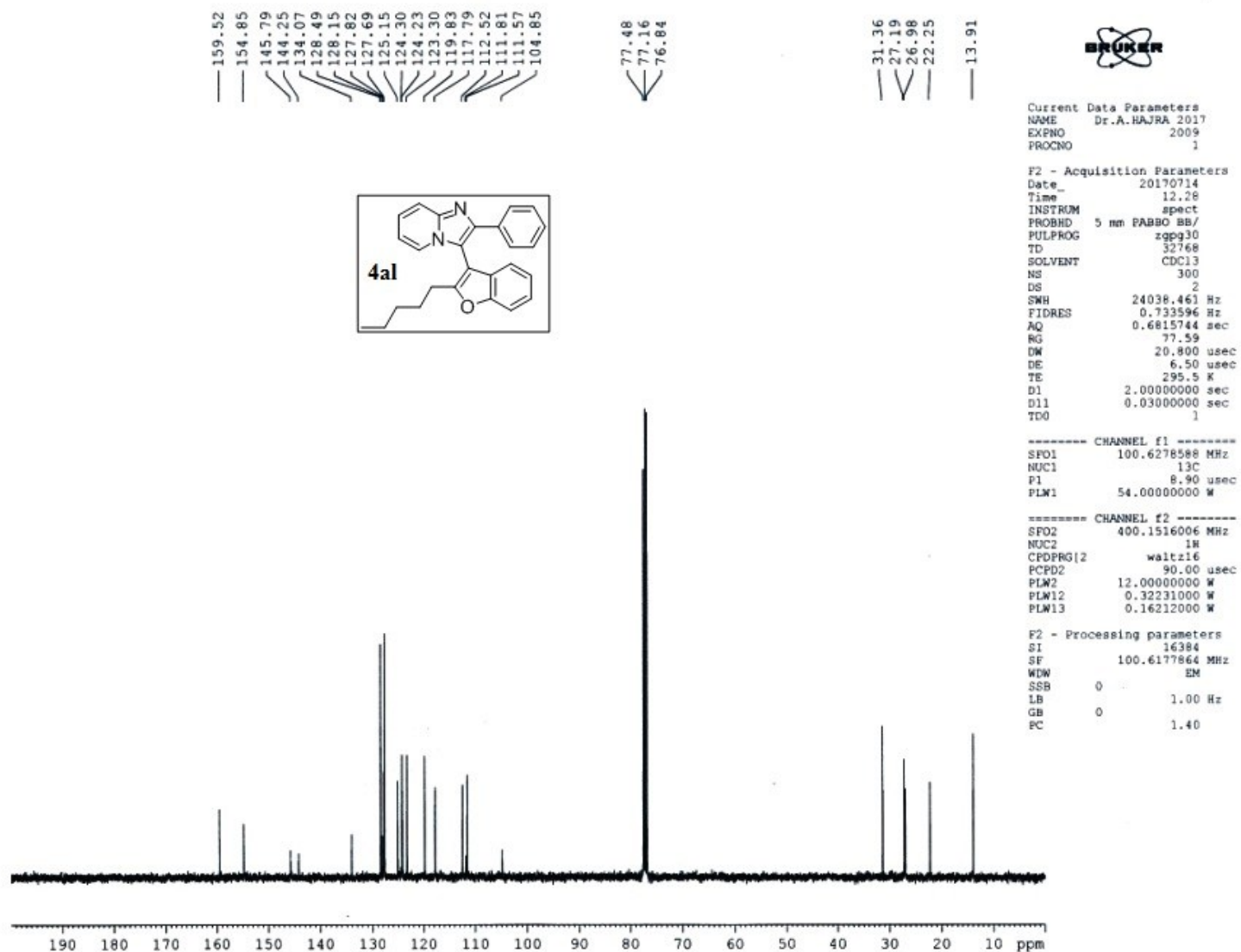


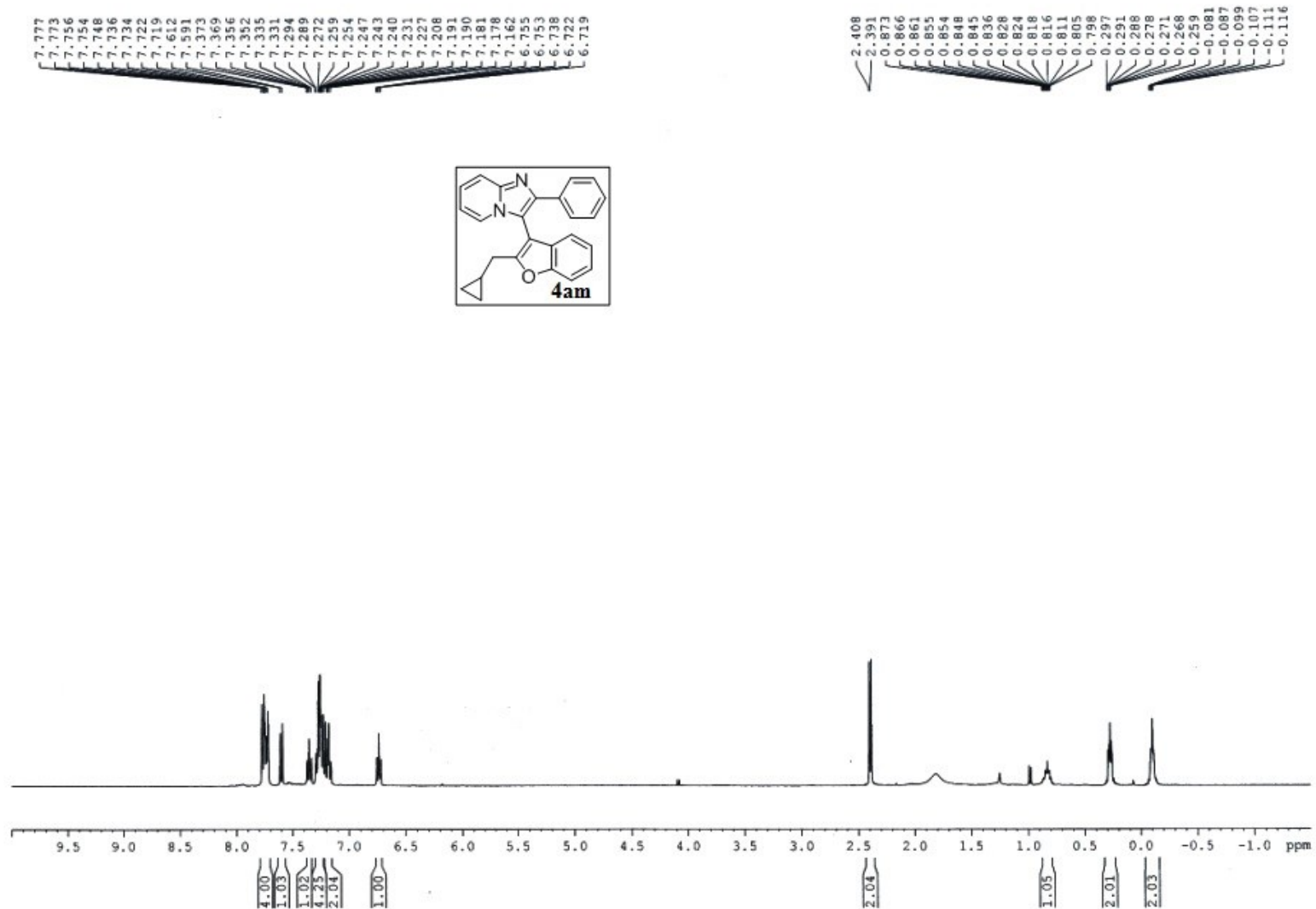


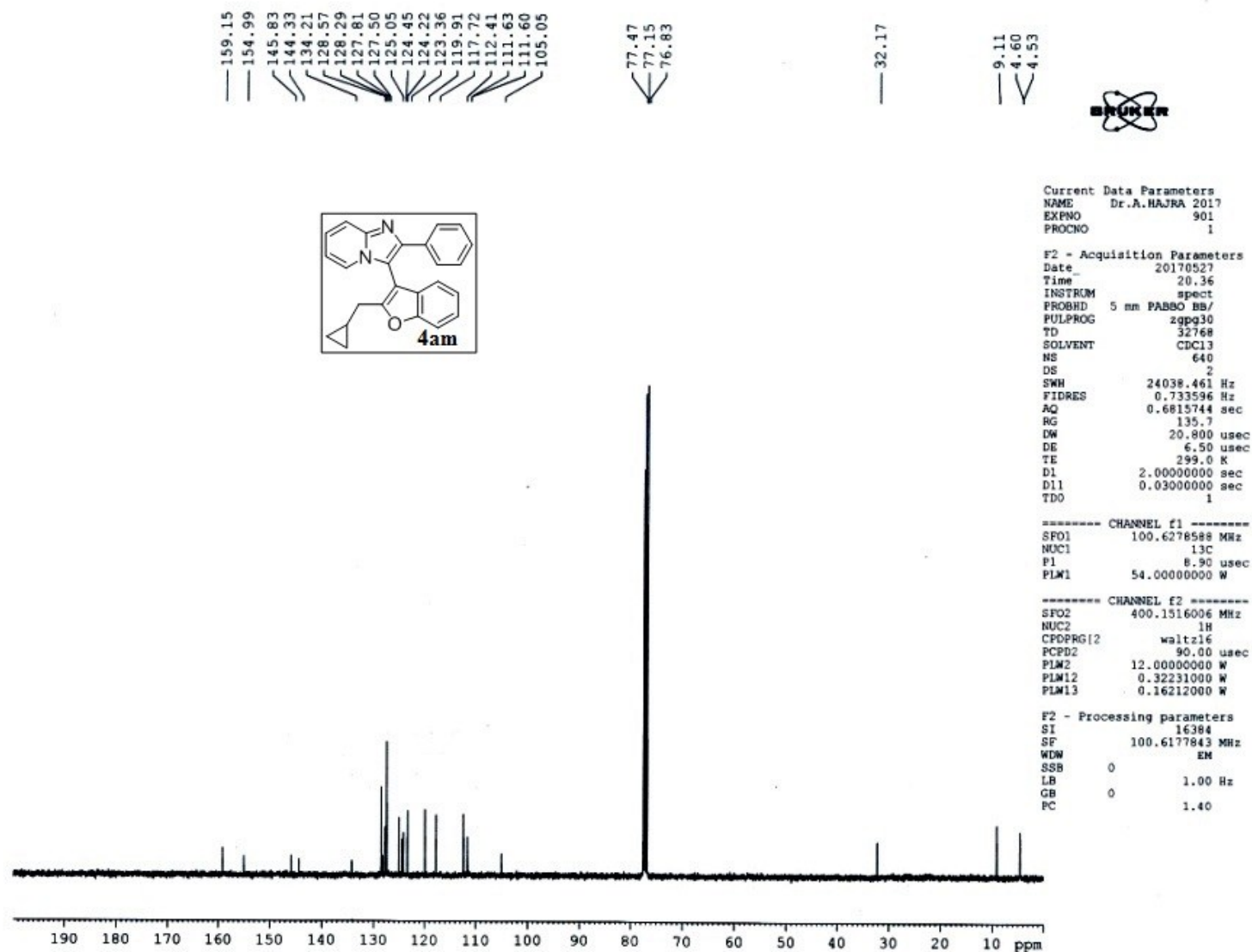


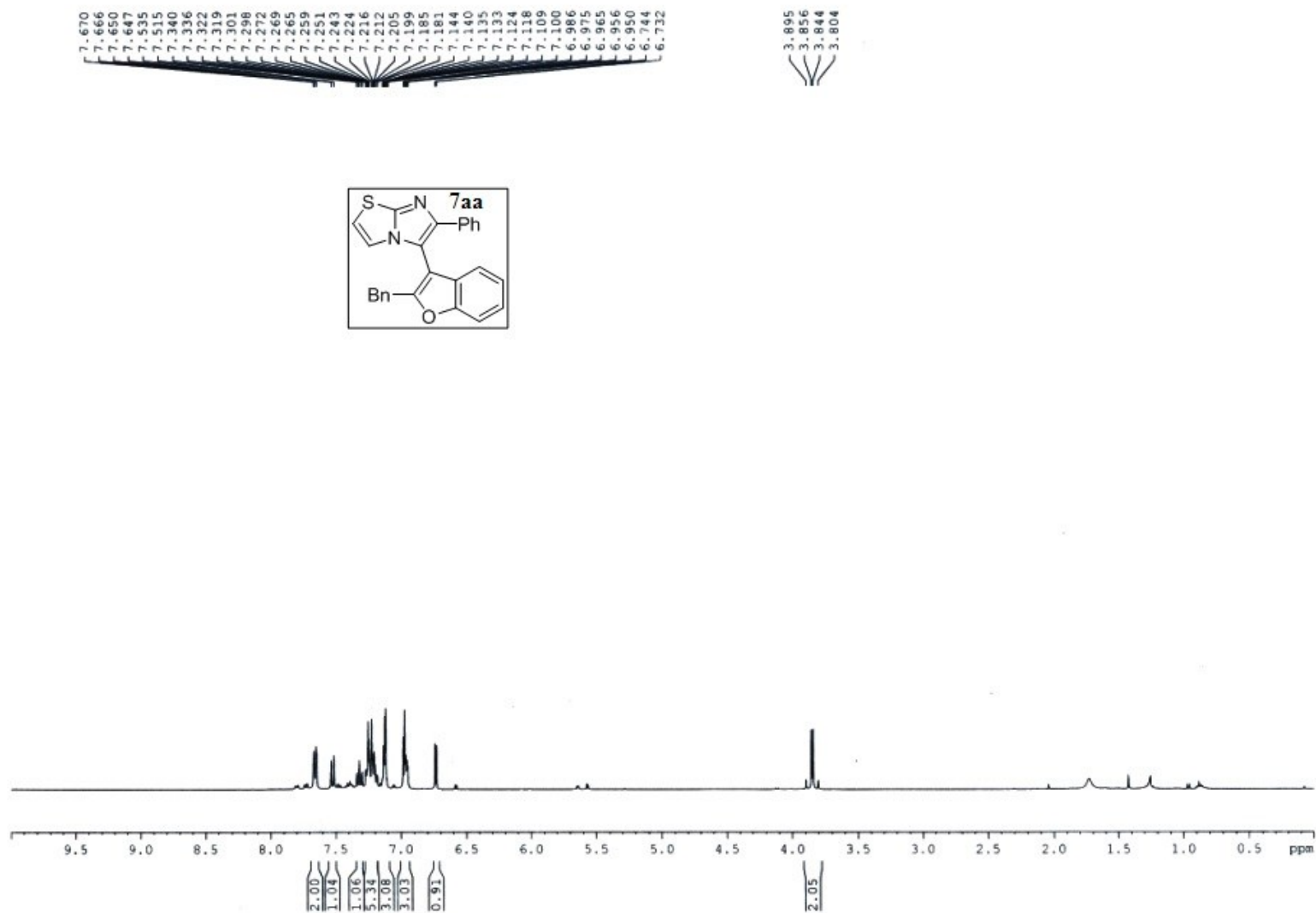


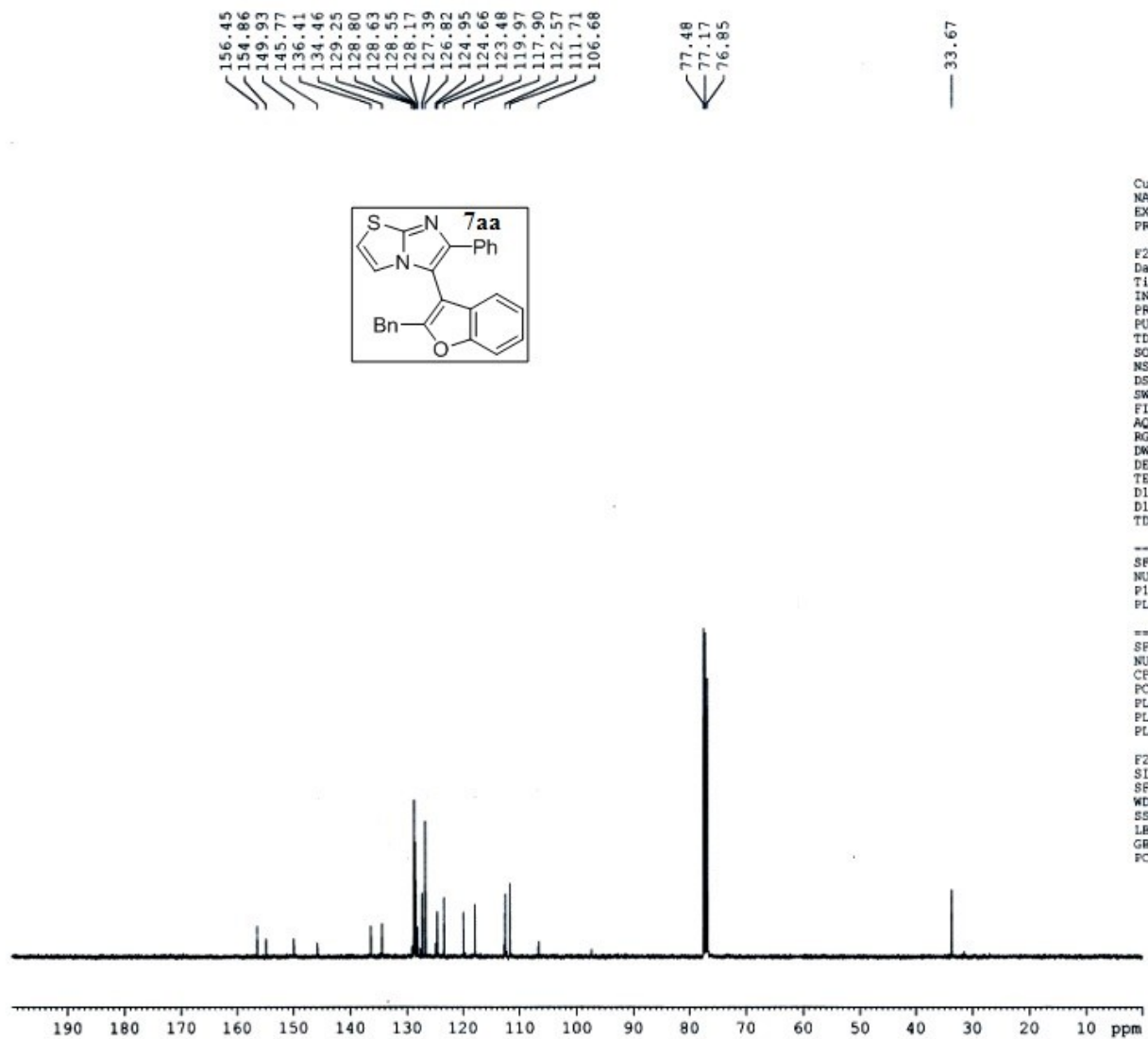












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Current Data Parameters
NAME      Dr.A.HAJRA 2017
EXPNO     1212
PROCNO    1

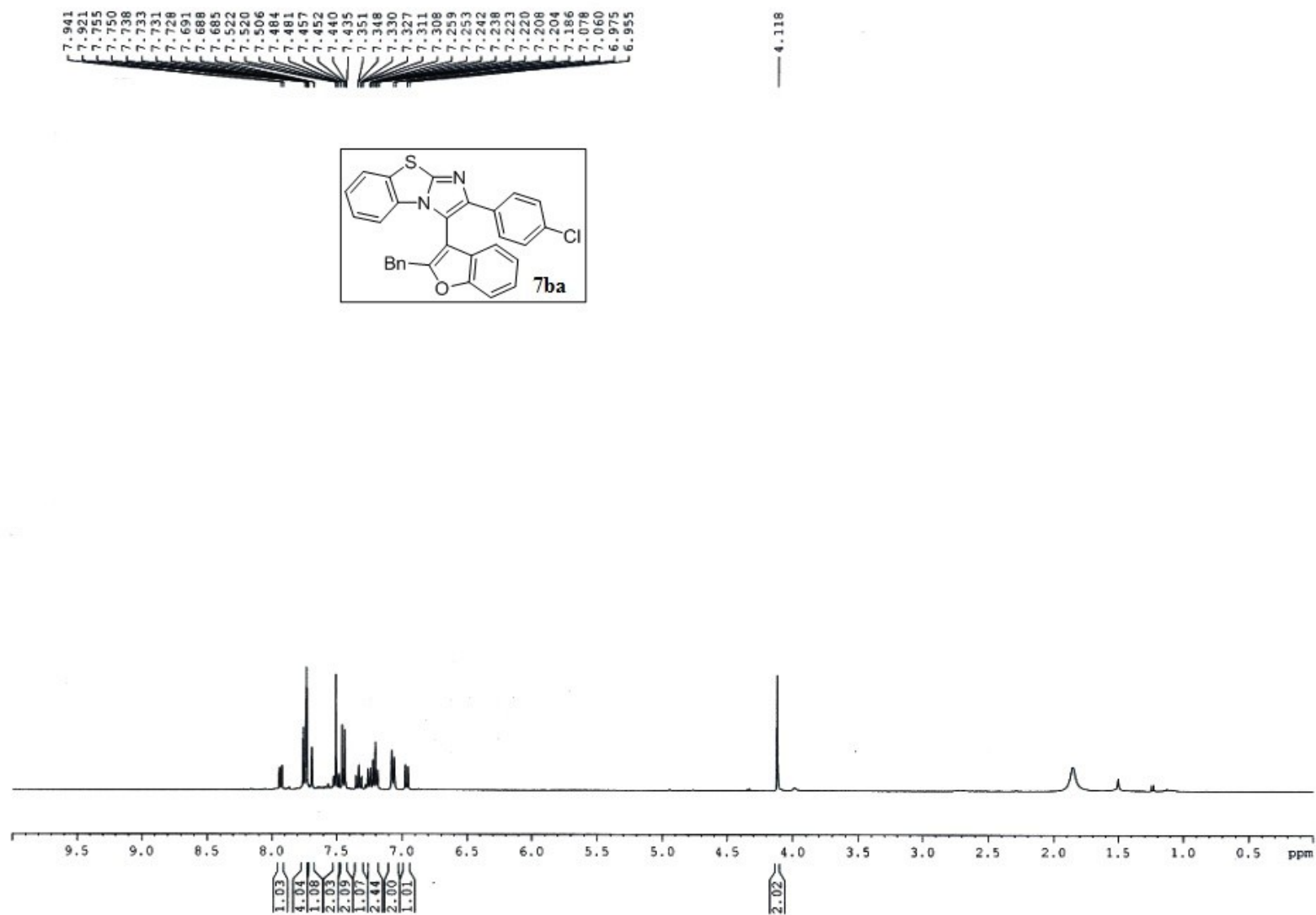
F2 - Acquisition Parameters
Date_     20170715
Time      21.39
INSTRUM   spect
PROBHD    5 mm PABBO BB/
PULPROG   zgpg30
TD         32768
SOLVENT    CDCl3
NS         640
DS         2
SWH        24038.461 Hz
FIDRES     0.733596 Hz
AQ         0.6815744 sec
RG         106.66
DW         20.800 usec
DE         6.50 usec
TE         298.6 K
D1         2.00000000 sec
D11        0.03000000 sec
TD0        1

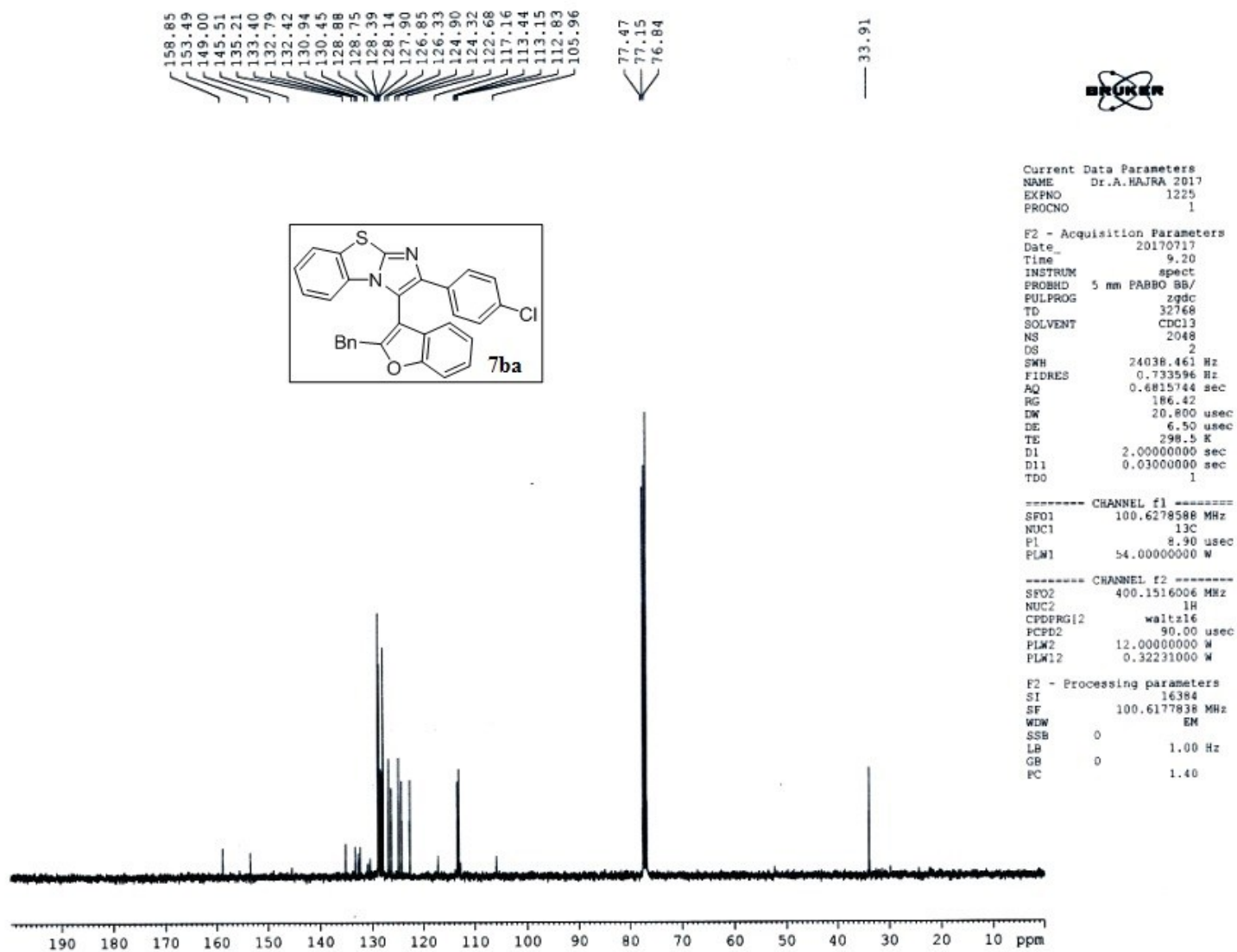
----- CHANNEL f1 -----
SFO1      100.6278588 MHz
NUC1       13C
P1         8.90 usec
PLW1       54.00000000 W

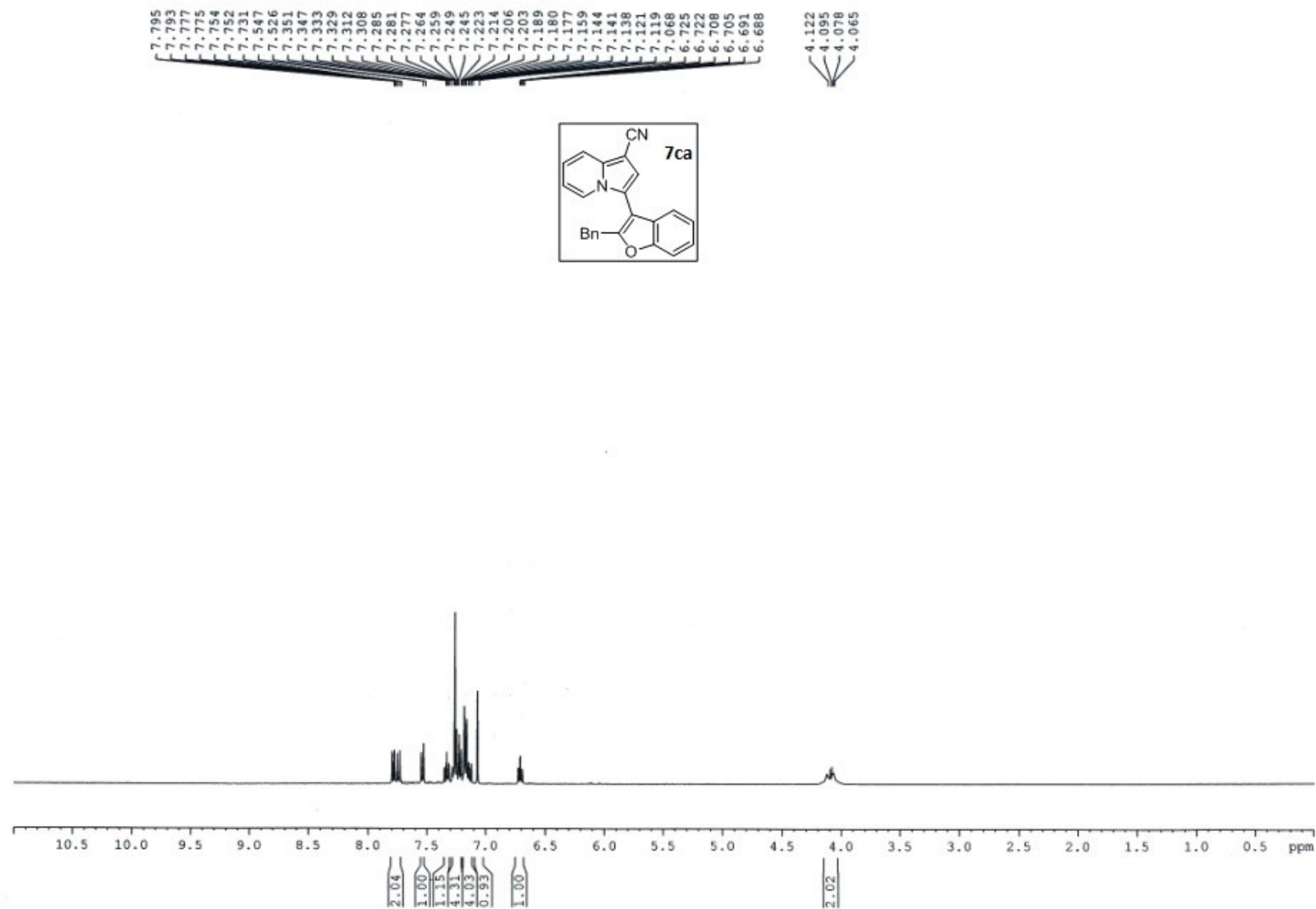
----- CHANNEL f2 -----
SFO2      400.1516006 MHz
NUC2       1H
CPDPRG2    waltz16
PCPD2      90.00 usec
PLW2       12.00000000 W
PLW12      0.32231000 W
PLW13      0.16212000 W

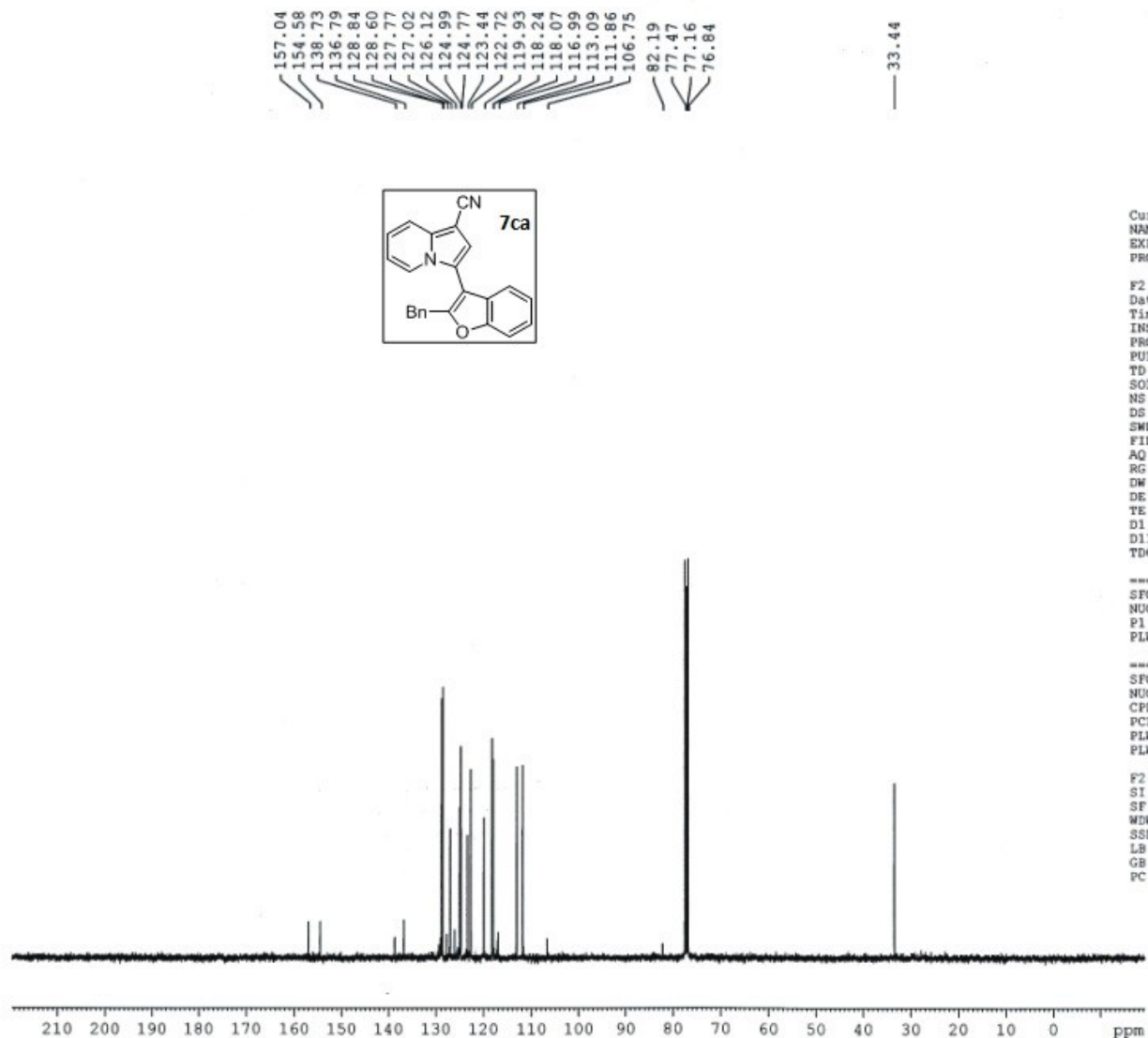
F2 - Processing parameters
SI         16384
SF         100.6177858 MHz
WDW        EM
SSB        0
LB         1.00 Hz
GB         0
PC         1.40

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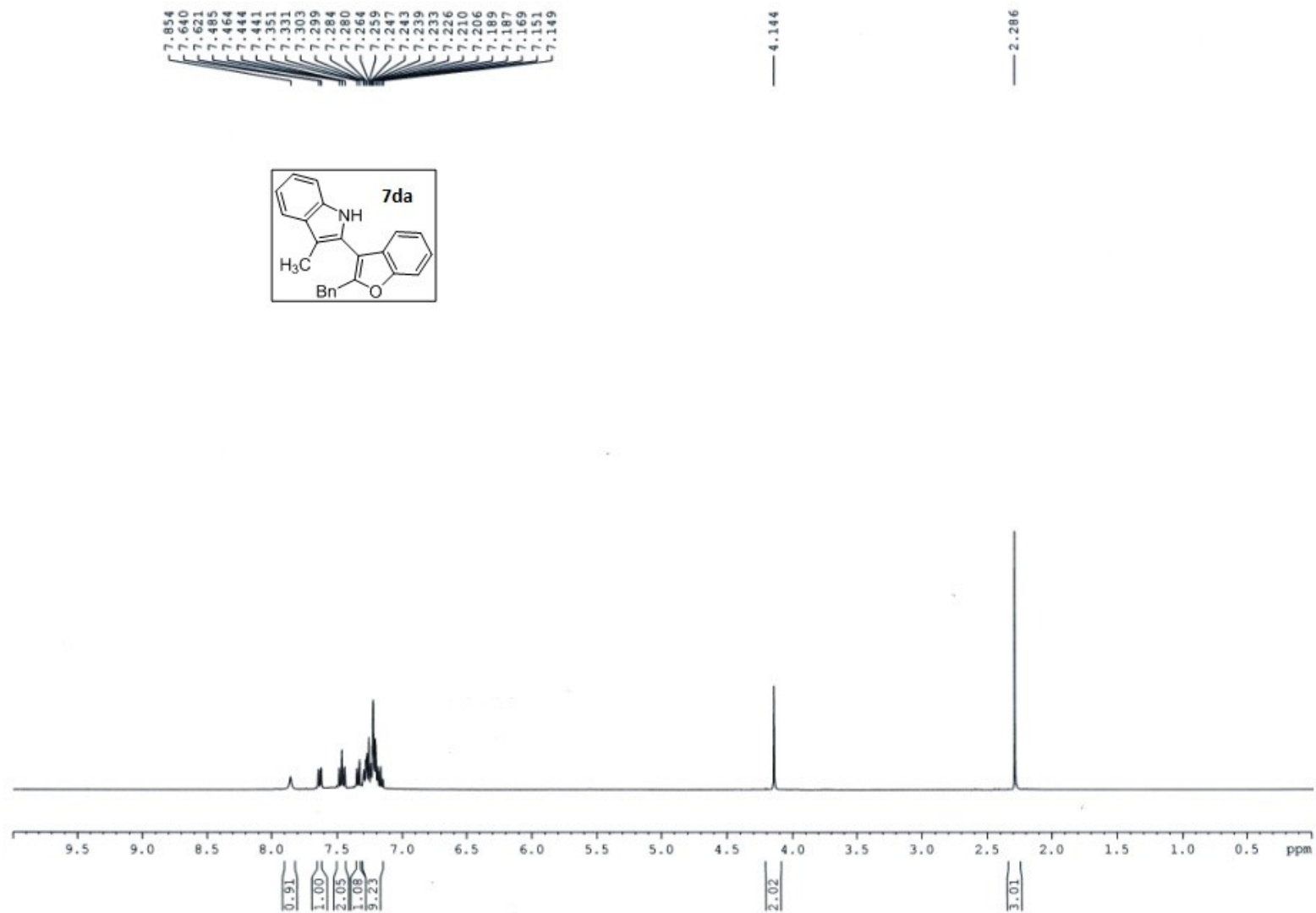
Current Data Parameters
 NAME Dr.A.HAJRA 2018
 EXPNO 345
 PROCNO 1

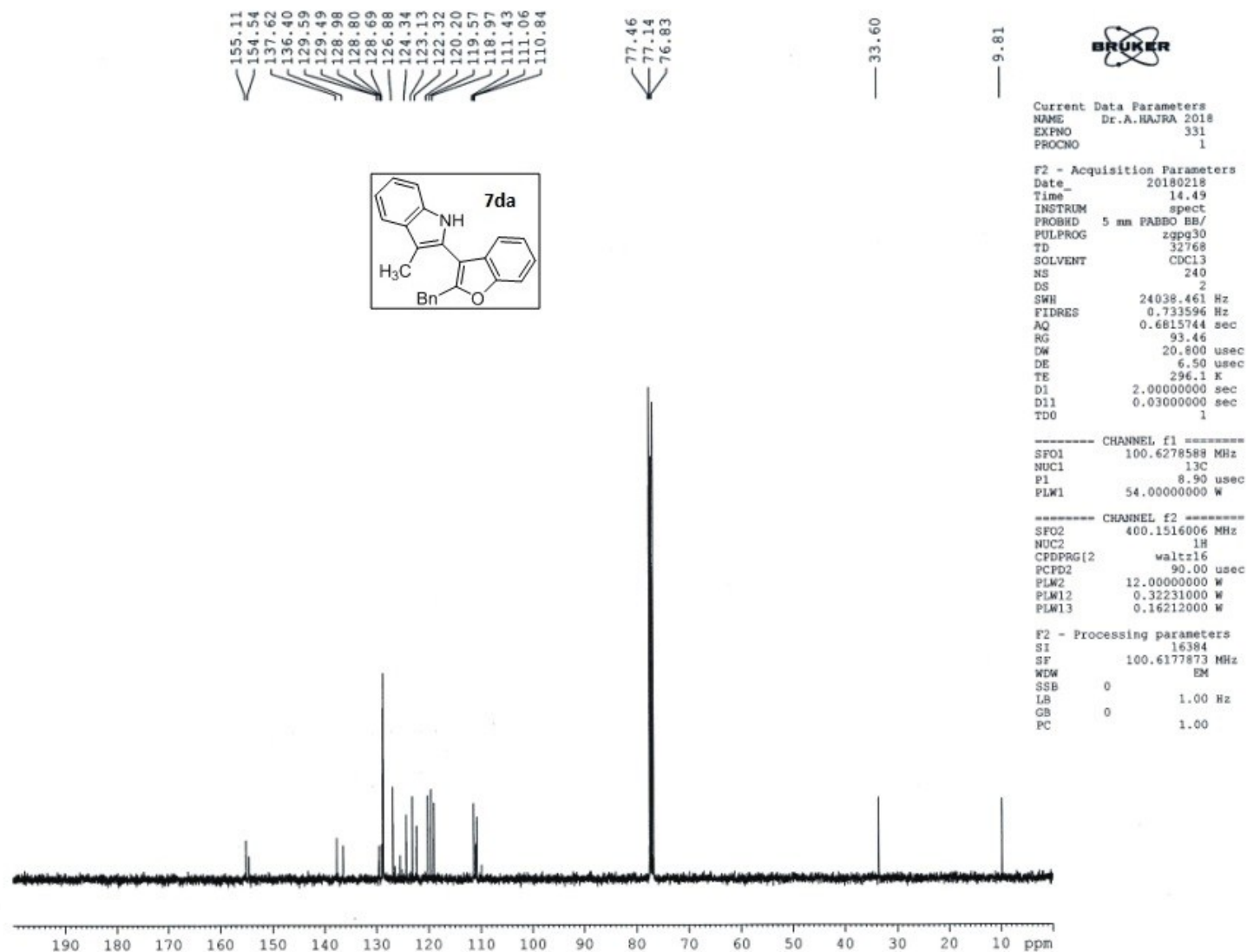
F2 - Acquisition Parameters
 Date_ 20180219
 Time 11.41
 INSTRUM spect
 PROBHD 5 mm PABBO BB/
 PULPROG zgpg
 TD 32768
 SOLVENT CDCl3
 NS 1024
 DS 2
 SWH 24038.461 Hz
 FIDRES 0.733596 Hz
 AQ 0.6815744 sec
 RG 186.42
 DW 20.800 usec
 DE 6.50 usec
 TE 294.3 K
 D1 2.00000000 sec
 D11 0.03000000 sec
 TDO 1

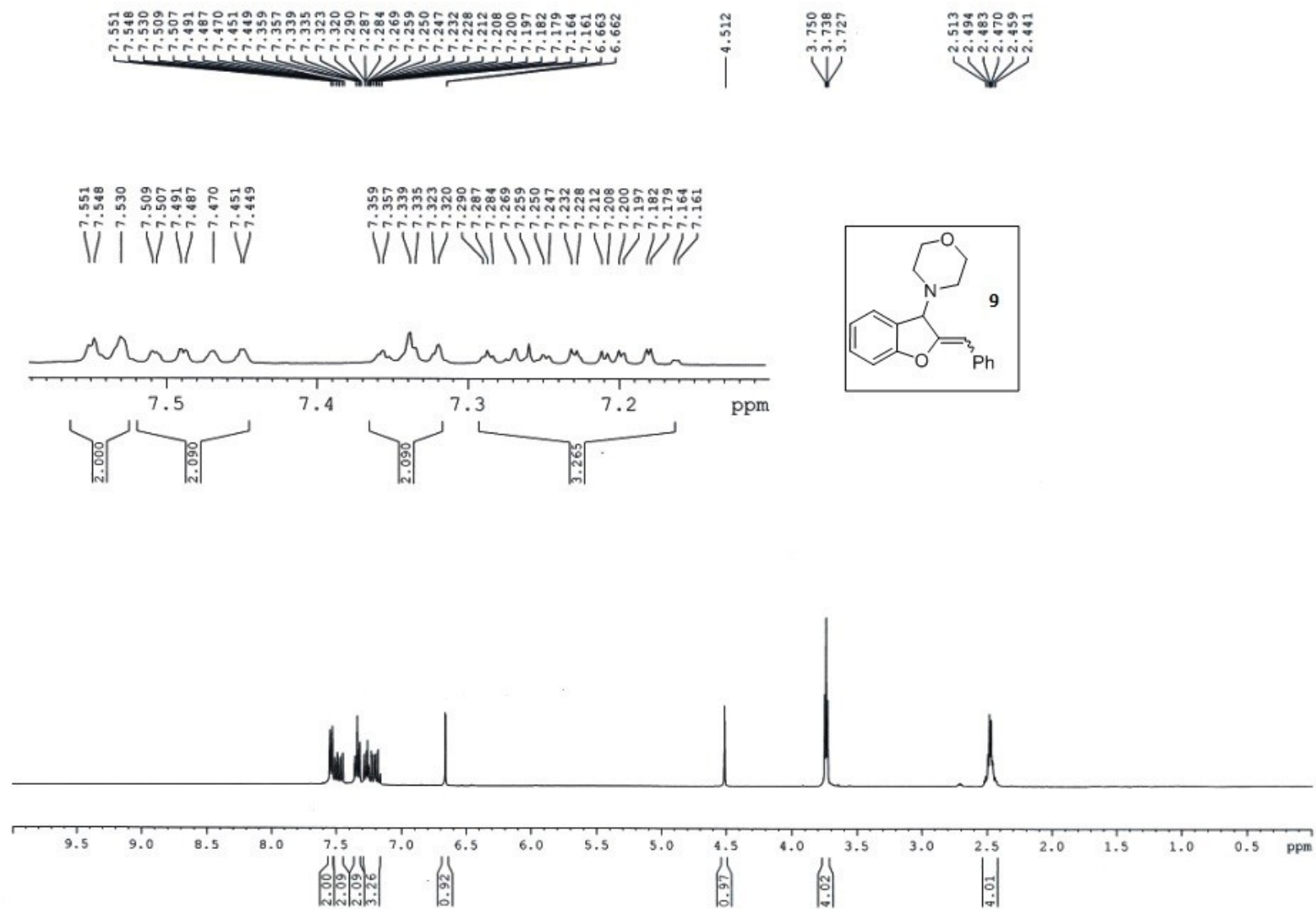
===== CHANNEL f1 =====
 SFO1 100.6278588 MHz
 NUC1 13C
 P1 8.90 usec
 PLW1 54.00000000 W

===== CHANNEL f2 =====
 SFO2 400.1516006 MHz
 NUC2 1H
 CPDPRG[2] waltz16
 PCPD2 90.00 usec
 PLW2 12.00000000 W
 PLW12 0.32231000 W

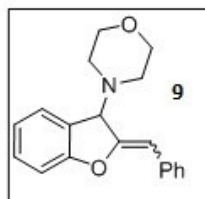
F2 - Processing parameters
 SI 16384
 SF 100.6177860 MHz
 WDW EM
 SSB 0
 LB 1.00 Hz
 GB 0
 PC 1.40







156.99
155.11
138.74
128.73
128.66
128.23
127.94
124.03
122.85
120.87
111.55
105.31
77.47
77.15
76.84
69.89
67.18
52.36



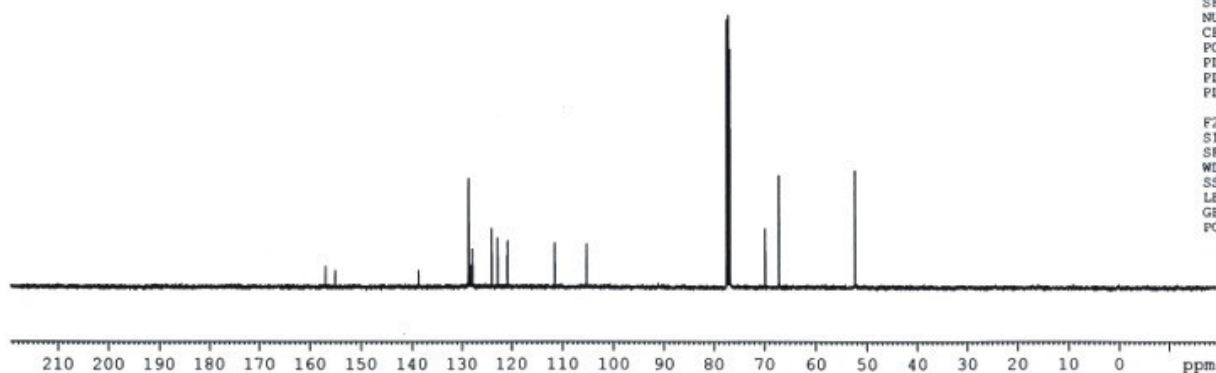
Current Data Parameters
NAME Dr.A.HAJRA 2018
EXPNO 257
PROCNO 1

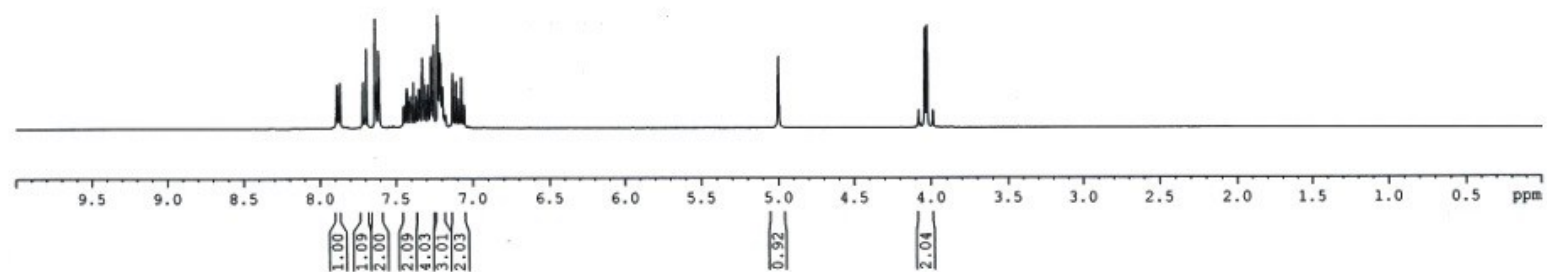
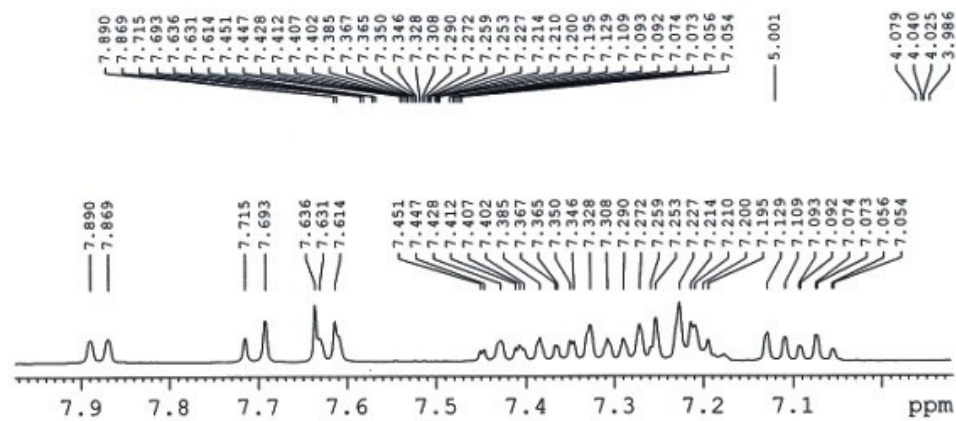
F2 - Acquisition Parameters
Date_ 20180215
Time 17.46
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDC13
NS 320
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 120.16
DW 20.800 usec
DE 6.50 usec
TE 296.0 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

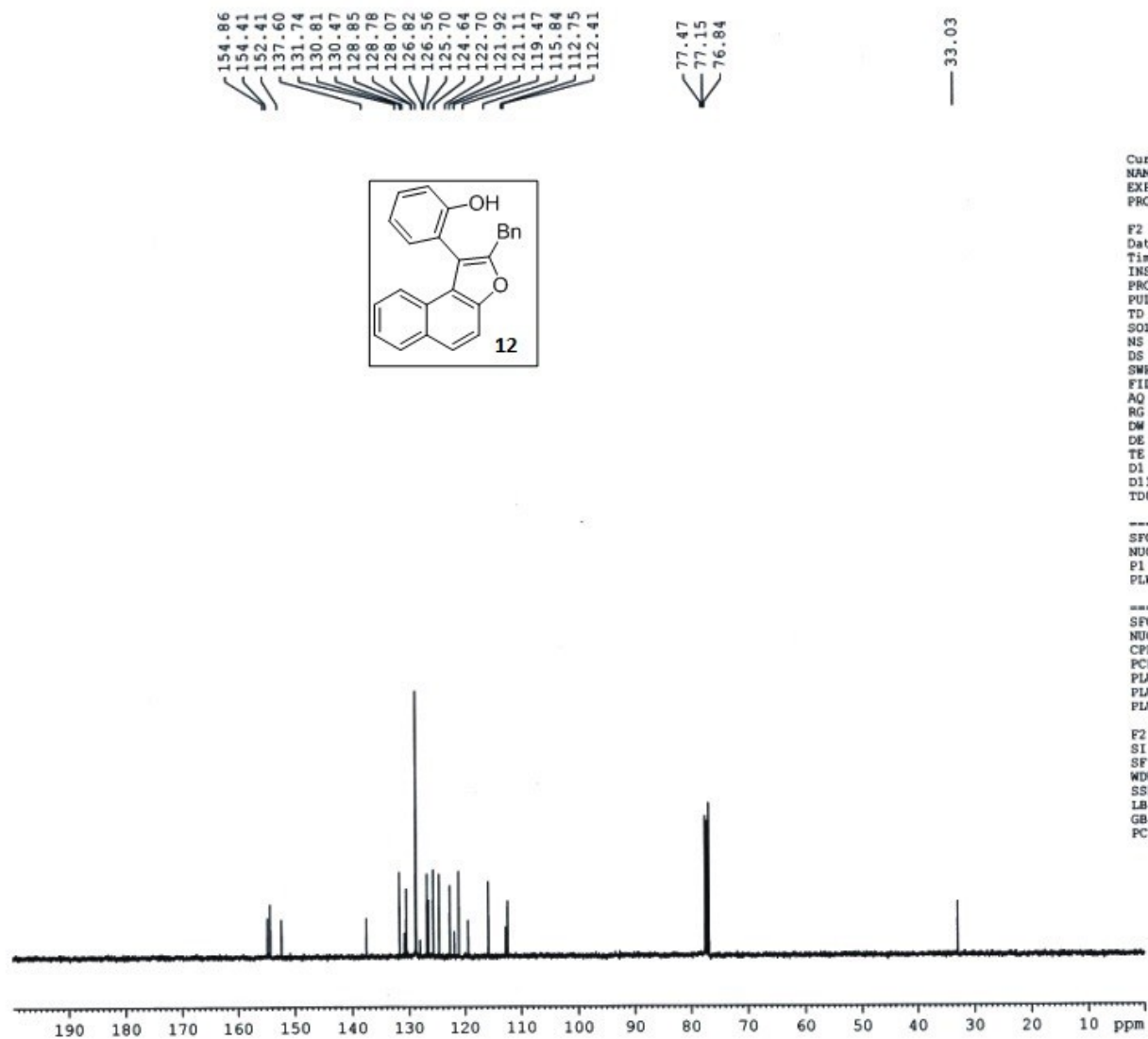
----- CHANNEL f1 -----
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

===== CHANNEL f2 =====
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG[2] waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

F2 - Processing parameters
SI 16384
SF 100.6177858 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.00







Current Data Parameters
NAME Dr.A.HAJRA 2017
EXPNO 1820
PROCNO 1

F2 - Acquisition Parameters
Date_ 20171104
Time_ 21.13
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 32768
SOLVENT CDCl3
NS 80
DS 2
SWH 24038.461 Hz
FIDRES 0.733596 Hz
AQ 0.6815744 sec
RG 62.69
CW 20.800 usec
DE 6.50 usec
TE 297.2 K
D1 2.00000000 sec
D11 0.03000000 sec
TD0 1

----- CHANNEL f1 -----
SFO1 100.6278588 MHz
NUC1 13C
P1 8.90 usec
PLW1 54.00000000 W

----- CHANNEL f2 -----
SFO2 400.1516006 MHz
NUC2 1H
CPDPRG12 waltz16
PCPD2 90.00 usec
PLW2 12.00000000 W
PLW12 0.32231000 W
PLW13 0.16212000 W

F2 - Processing parameters
SI 16384
SF 100.6177897 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.20