

## Supporting Information

### **Copper(II) Complex Containing Pyridine-2-Carbaldehyde and its Direct Binding onto Ethylenediamine Functionalized with Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub> Nanoparticles for the Catalytic Applications**

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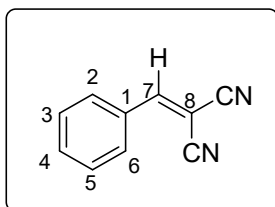
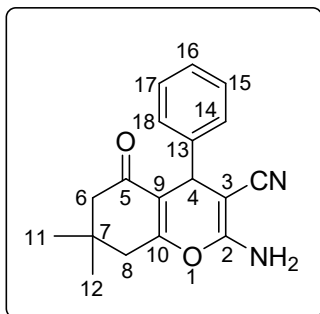
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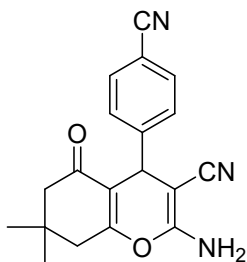
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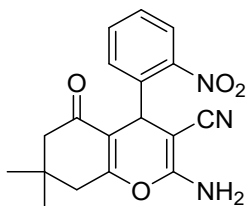




**2-Amino-7,7-dimethyl-4-(4-cyanophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran**

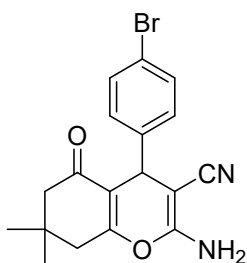
FT-IR (KBr,  $\text{cm}^{-1}$ ): 3354, 3062, 2962, 2227, 2190, 1693, 1503, 1470, 1039, 59, 74.  $^1\text{H-NMR}$  (250 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  7.72 (d, 2H,  $J = 7$ , Ar-H), 7.33 (d, 2H,  $J = 7.25$ , Ar-H), 7.10 (s, 2H,  $\text{NH}_2$ ), 4.26 (s, 1H, C-H), 2.19-2.25 (m, 2H,  $\text{CH}_2$ ), 2.04-2.10 (m, 2H,  $\text{CH}_2$ ), 0.99 (s, 3H, Me), 0.91 (s, 3H, Me).  $^{13}\text{C-NMR}$  (62.5 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  27.3 (C11 & C12), 28.6 (C7), 32.2 (C8), 36.2 (C4), 50.2 (C6), 56.4 (C3), 109.8 (C16), 112.1 (C9), 119.1 (CN), 119.8 (CN), 128.7 (C18 & C14), 132.8 (C15 & C17), 150.6 (C13), 159.5 (C10), 163.5 (C2), 196.1 (C5).

**2-Amino-7, 7-dimethyl-4-(2-nitrophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran**



FT-IR (KBr,  $\text{cm}^{-1}$ ): 3333, 3185, 2957, 2193, 1694, 1580, 1526, 1469, 1327, 1255, 1041, 869, 736.  $^1\text{H-NMR}$  (250 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  7.76 (d, 1H,  $J = 7$ , Ar-H), 7.59 (d, 1H,  $J = 7.25$ , Ar-H), 7.29-7.39 (m, 2H, Ar-H), 7.16 (s, 2H,  $\text{NH}_2$ ), 4.88 (s, 1H, C-H), 2.12-2.18 (m, 2H,  $\text{CH}_2$ ), 1.92-1.99 (m, 2H,  $\text{CH}_2$ ), 0.96 (s, 3H, Me), 0.82 (s, 3H, Me).  $^{13}\text{C-NMR}$  (62.5 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  27.0 (C11 & C12), 28.6 (C7), 30.3 (C8), 32.2 (C4), 49.9 (C6), 56.7 (C3), 112.7 (C9), 119.4 (CN), 124.1 (C15), 128.2 (C16), 130.6 (C18), 133.6 (C17), 133.7 (C13), 139.3 (C14), 149.4 (C10), 159.5 (C2), 196.2 (C5).

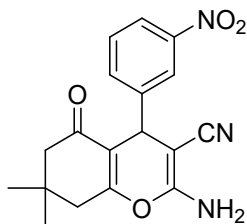
**2-Amino-7, 7-dimethyl-4-(4-bromophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran**



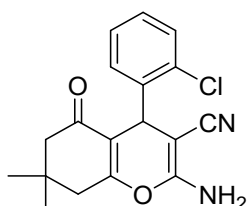
FT-IR (KBr,  $\text{cm}^{-1}$ ): 3364, 3153, 2965, 2191, 166, 1607, 1473, 1365, 1255, 1042, 632.  $^1\text{H-NMR}$  (250 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  7.45 (d, 2H,  $J = 7.25$ , Ar-H), 7.07 (d, 2H,  $J = 8.5$ , Ar-H), 7.04 (s, 2H,  $\text{NH}_2$ ), 4.14 (s, 1H, C-H), 2.18-2.25 (m, 2H,  $\text{CH}_2$ ), 2.03-2.09 (m, 2H,  $\text{CH}_2$ ), 1.0 (s, 3H, Me), 0.91 (s, 3H, Me).  $^{13}\text{C-NMR}$  (62.5 MHz,  $\text{DMSO-d}_6$ ):  $\delta$  27.2 (C11 & C12), 28.7 (C7), 32.2 (C8), 35.6 (C4), 50.3 (C6), 58.0 (C3), 112.7 (C9), 119.9 (CN), 129.9 (C15 & C17), 131.6 (C14 & C18), 144.6 (C13), 158.9 (C10), 163.0 (C2), 196.0 (C5).

**2-Amino-7, 7-dimethyl-4-(3-nitrophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran**

FT-IR (KBr,  $\text{cm}^{-1}$ ): 3430, 3335, 3100, 2956, 2186, 1685, 1599, 1529, 1417, 1347, 1427, 1252, 1037, 714.  $^1\text{H-NMR}$  (250 MHz,  $\text{DMSO-d}_6$ ):

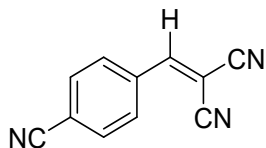


$\delta$  7.61-8.03 (m, 4H, Ar-H), 7.16 (s, 2H, NH<sub>2</sub>), 4.38 (s, 1H, CH), 2.04-2.51 (m, 4H, 2CH<sub>2</sub>), 1.0 (s, 3H, Me), 0.91 (s, 3H, Me). <sup>13</sup>C-NMR (62.5 MHz, DMSO-d<sub>6</sub>):  $\delta$  27.1 (C11 & C12), 28.7 (C7), 32.2 (C8), 35.8 (C4), 50.2 (C6), 57.5 (C3), 112.2 (C9), 119.8 (CN), 122.2 (C16), 130.4 (17), 134.6 (C18), 147.4 (C13), 148.2 (15), 159.0 (C10), 163.6 (C2), 196.2 (C5).



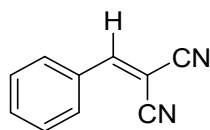
**2-Amino-7, 7-dimethyl-4-(2-chlorophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran**

FT-IR (KBr, cm<sup>-1</sup>): 3396, 3330, 3198, 2959, 2199, 1682, 1603, 1468, 1368, 1214, 1037, 750. <sup>1</sup>H-NMR (250 MHz, DMSO-d<sub>6</sub>):  $\delta$  7.14-7.31 (m, 4H, Ar-H), 7.01 (s, 2H, NH<sub>2</sub>), 4.65 (s, 1H, CH), 2.0-2.29 (m, 4H, 2CH<sub>2</sub>), 1.0 (s, 3H, Me), 0.94 (s, 3H, Me). <sup>13</sup>C-NMR (62.5 MHz, DMSO-d<sub>6</sub>):  $\delta$  27.3 (C11 & C12), 28.8 (C7), 32.1 (C8), 32.2 (C4), 50.3 (C6), 57.2 (C3), 112.2 (C9), 119.6 (CN), 127.8 (C18), 128.6 (C17), 129.8 (C16), 130.3 (C14), 132.5 (C13), 141.9 (C10), 159.1 (C2), 163.5 (C5).



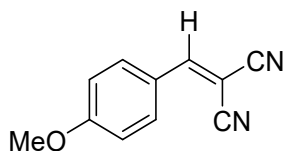
**2-(4-Cyanobenzylidene)malononitrile**

FT-IR (KBr, cm<sup>-1</sup>): 3044, 2307, 2230, 1652, 1414, 1211, 960, 785, 619. <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>):  $\delta$  7.99 (d, 2H, *J* = 7, Ar-H), 7.82 (d, 3H, *J* = 3.5, Ar-H, C-H). <sup>13</sup>C NMR (62.5 MHz, CDCl<sub>3</sub>):  $\delta$  111.7 (C4), 112.7 (2CN), 117.3 (CN), 130.7 (C2 & C6), 133.1 (C3 & C5), 134.2 (C1), 157.4 (C7).



**2-Benzylidenemalononitrile**

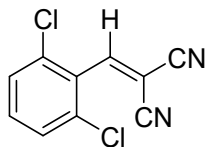
FT-IR (KBr, cm<sup>-1</sup>): 3012, 2223, 1592, 1449, 1217, 957, 775. <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>):  $\delta$  7.23-7.54 (m, 3H, Ar-H), 7.76 (s, 1H, C-H), 7.88 (m, 2H, Ar-H). <sup>13</sup>C NMR (62.5 MHz, CDCl<sub>3</sub>):  $\delta$  82.8 (C8), 112.5 (CN), 113.6 (CN), 129.6 (C4, C3 & C5), 130.7 (C2 & C6), 134.6 (C1), 159.9 (C7).



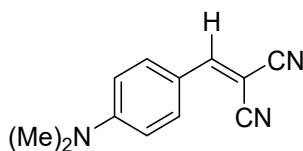
**2-(4-Methoxybenzylidene)malononitrile**

FT-IR (KBr, cm<sup>-1</sup>): 2223, 1605, 1445, 1278, 1185, 833, 578. <sup>1</sup>H NMR (250 MHz, CDCl<sub>3</sub>):  $\delta$  7.63 (d, 2H, *J* = 8.25, Ar-H), 7.69 (s, 1H), 6.99 (d, 2H, *J* = 8.25, Ar-H), 3.89 (s, 3H, OMe). <sup>13</sup>C NMR (62.5 MHz, CDCl<sub>3</sub>):  $\delta$  55.8 (OMe), 78.3 (C8), 113.4 (CN), 114.4 (CN), 115.1 (C3 & C5), 124 (C1), 133.4 (C2 & C6), 158.9 (C4), 164.8 (C7).

### 2-(2,6-Dichlorobenzylidene)malononitrile

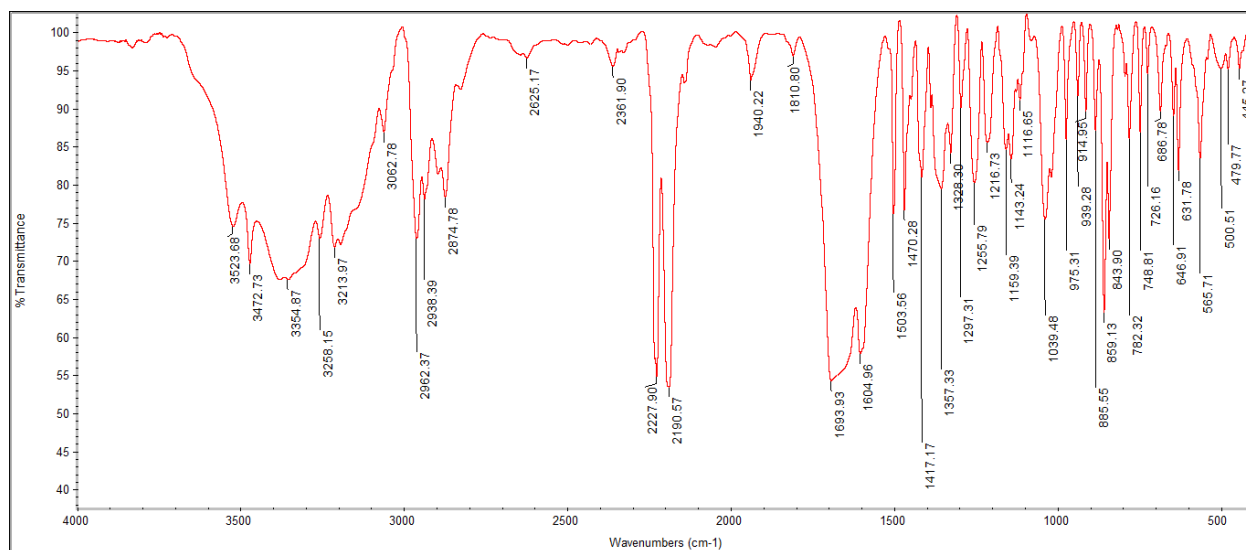


FT-IR (KBr,  $\text{cm}^{-1}$ ): 3026, 2236, 1610, 1432, 1197, 1098, 892, 615, 520.  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.94 (s, 1H, C-H), 7.37-7.42 (m, 3H, Ar-H).  $^{13}\text{C}$  NMR (62.5 MHz,  $\text{CDCl}_3$ ):  $\delta$  94.2 (C8), 110.6 (CN), 111.8 (CN), 128.8 (C4), 129.7 (C3 & C5), 132.8 (C2 & C6), 133.6 (C1), 156.6 (C7).



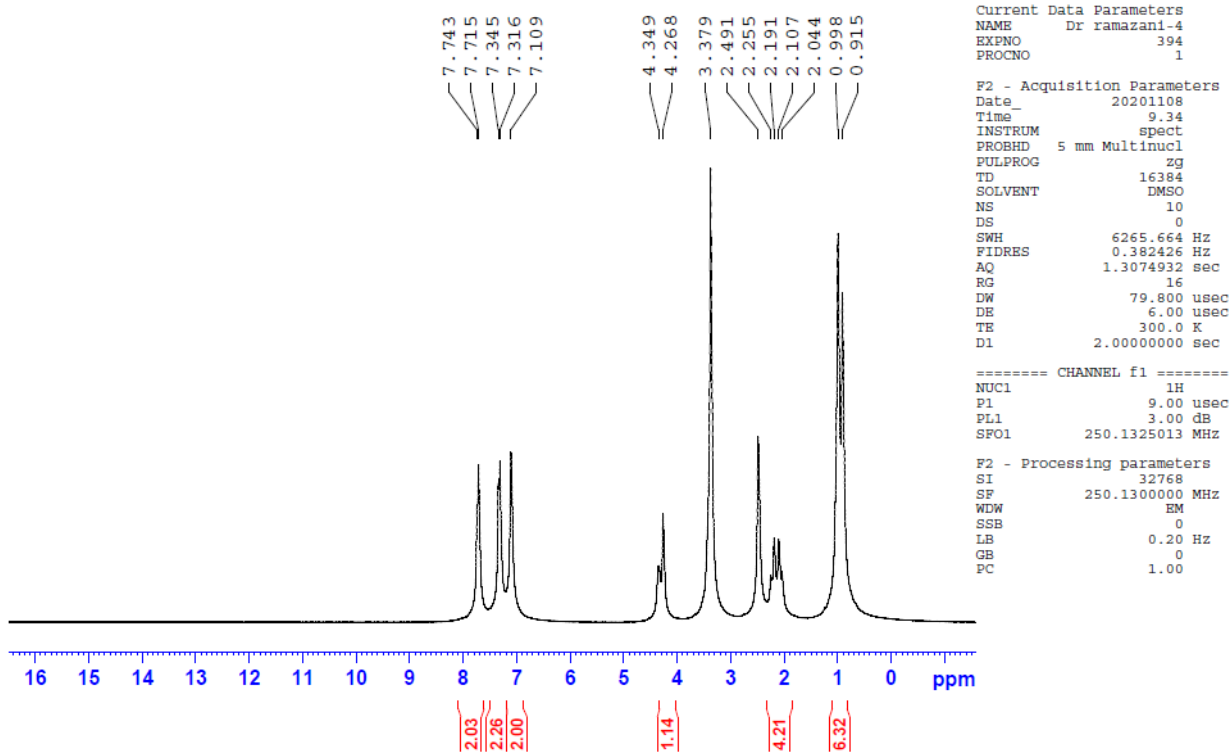
### 2-(4-(Dimethylamino)benzylidene)malononitrile

FT-IR (KBr,  $\text{cm}^{-1}$ ): 2210, 1614, 1521, 1198, 816, 727, 600, 518.  $^1\text{H}$  NMR (250 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.75 (d, 2H,  $J = 7$ , Ar-H), 7.38 (s, 1H, C-H), 6.66 (d, 2H,  $J = 7.25$ , Ar-H).  $^{13}\text{C}$  NMR (62.5 MHz,  $\text{CDCl}_3$ ):  $\delta$  (ppm): 40 (NMe<sub>2</sub>), 71.54 (C7), 111.6 (C3 & C5), 114.9 (CN), 116 (CN), 119.2 (C1), 133.8 (C2 & C6), 154.2 (C4) 158 (C7).

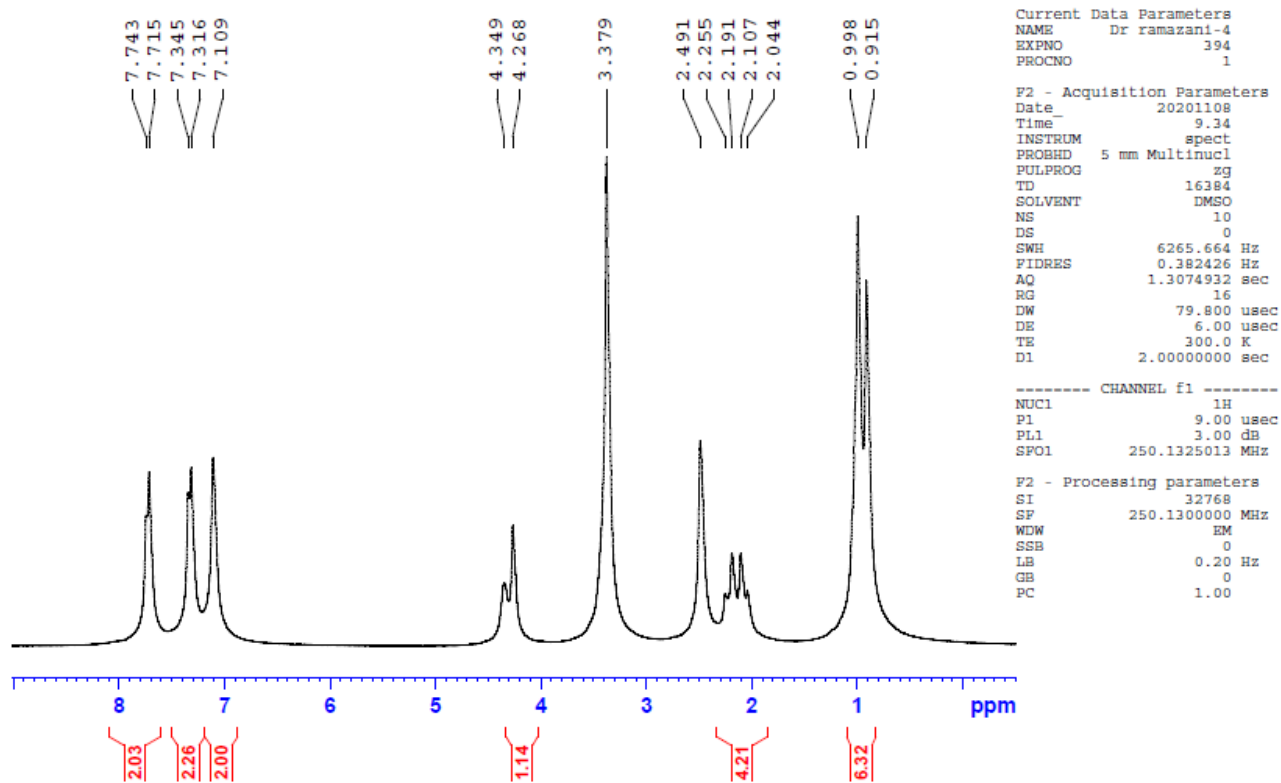


**Figure S1:** FT-IR spectrum of 2-amino-7,7-dimethyl-4-(4-cyanophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran

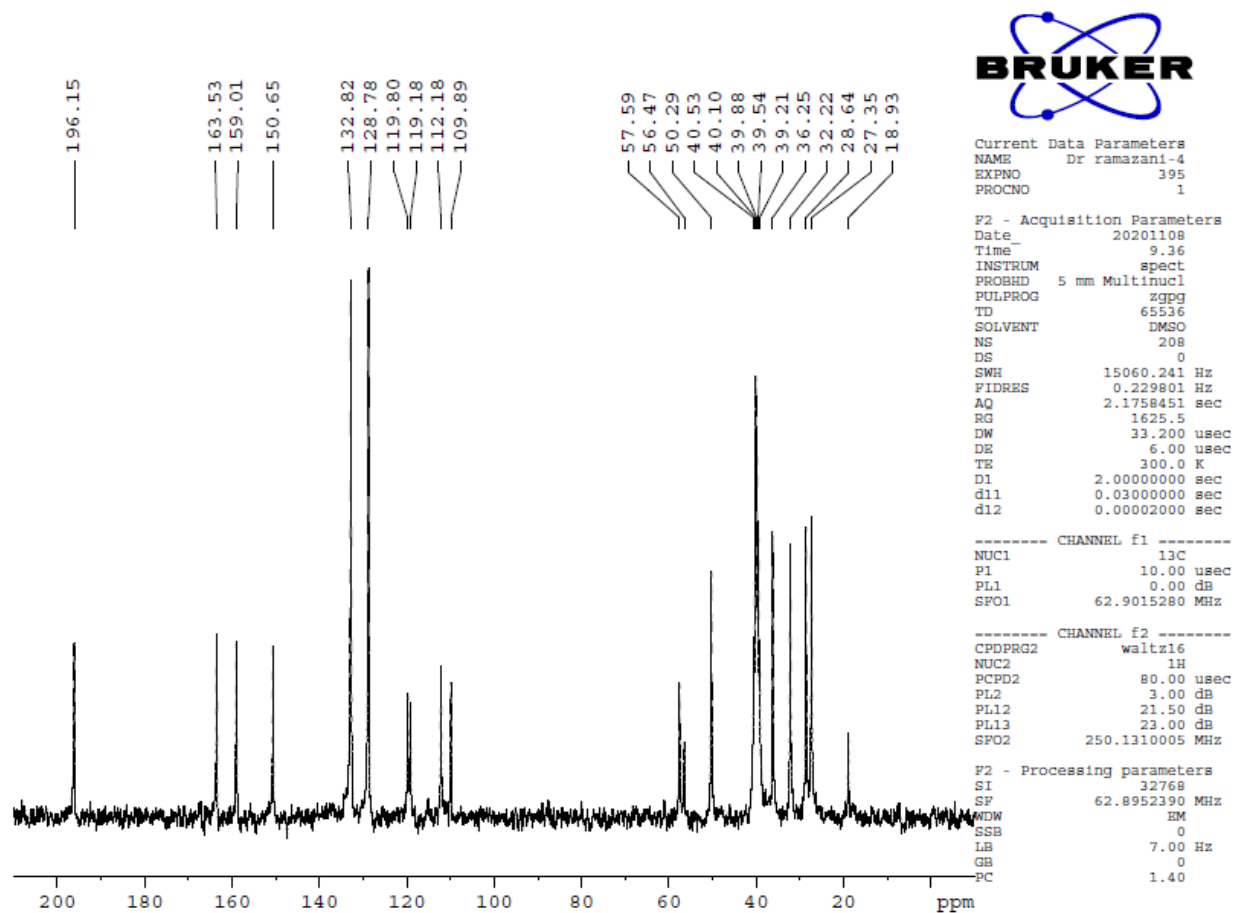




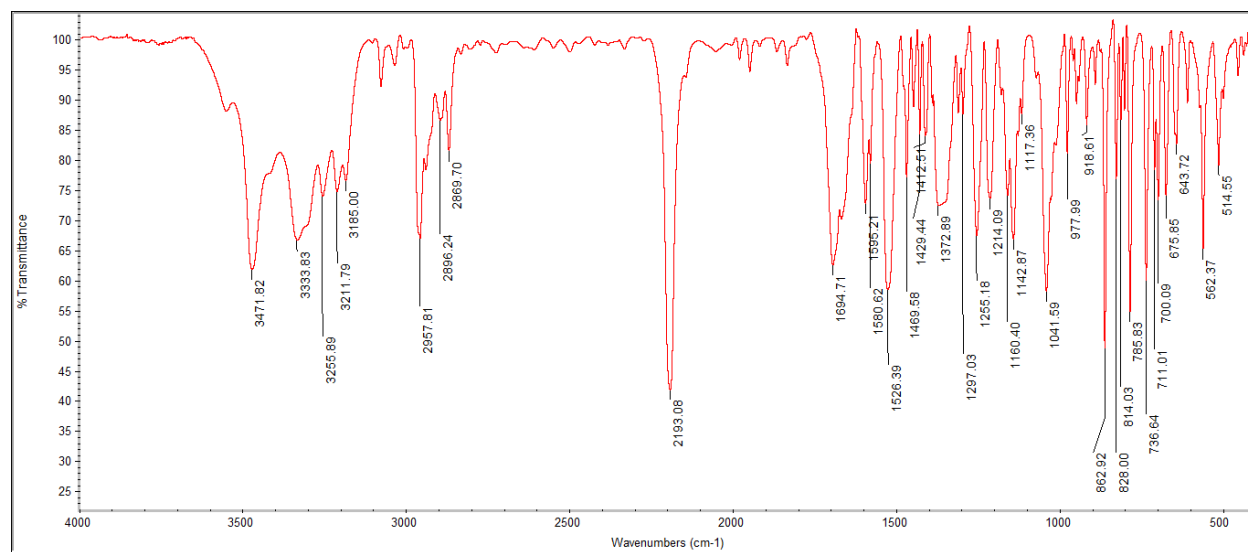
**Figure S2:**  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(4-cyanophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran



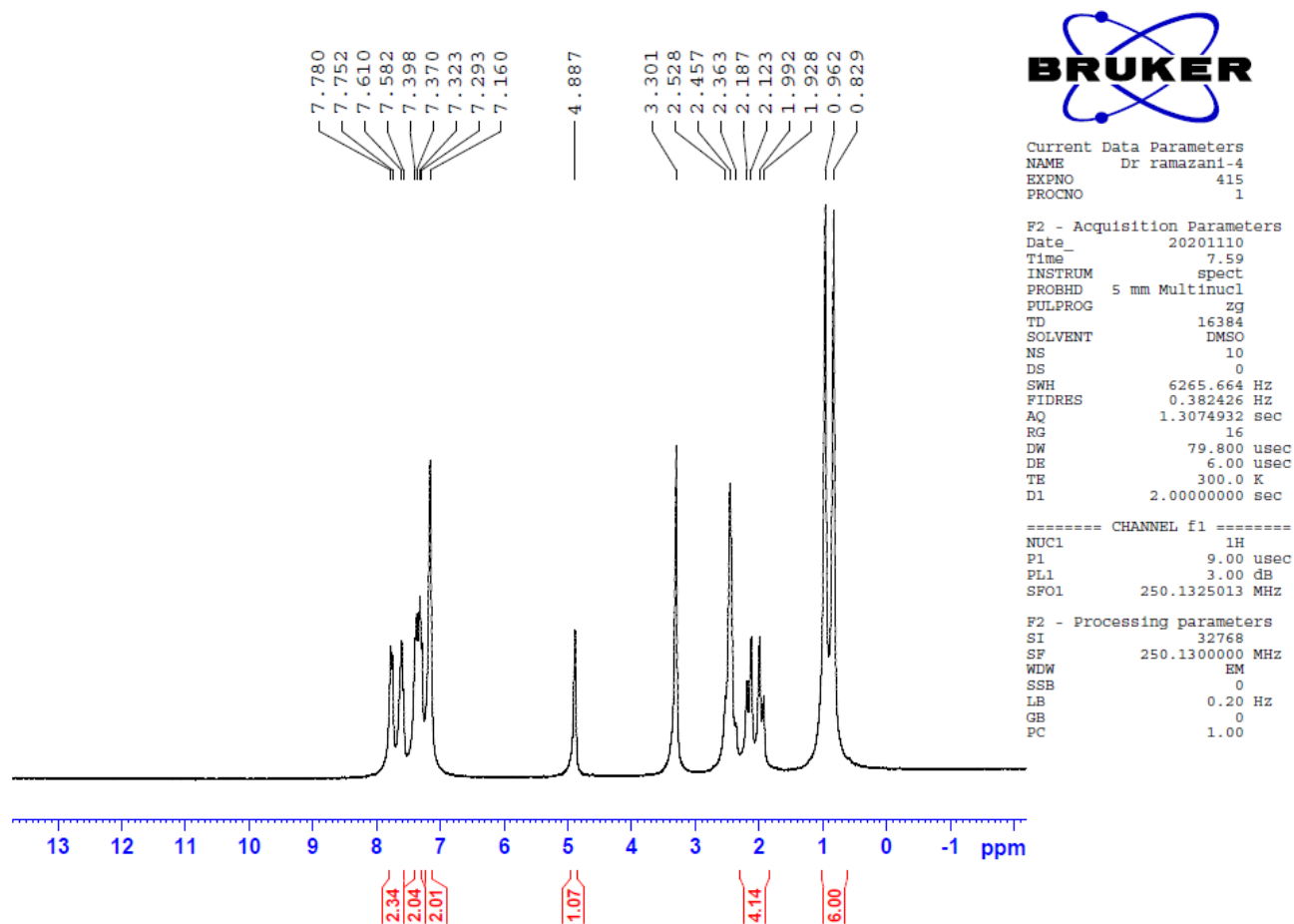
**Figure S3:** The expanded  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(4-cyanophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



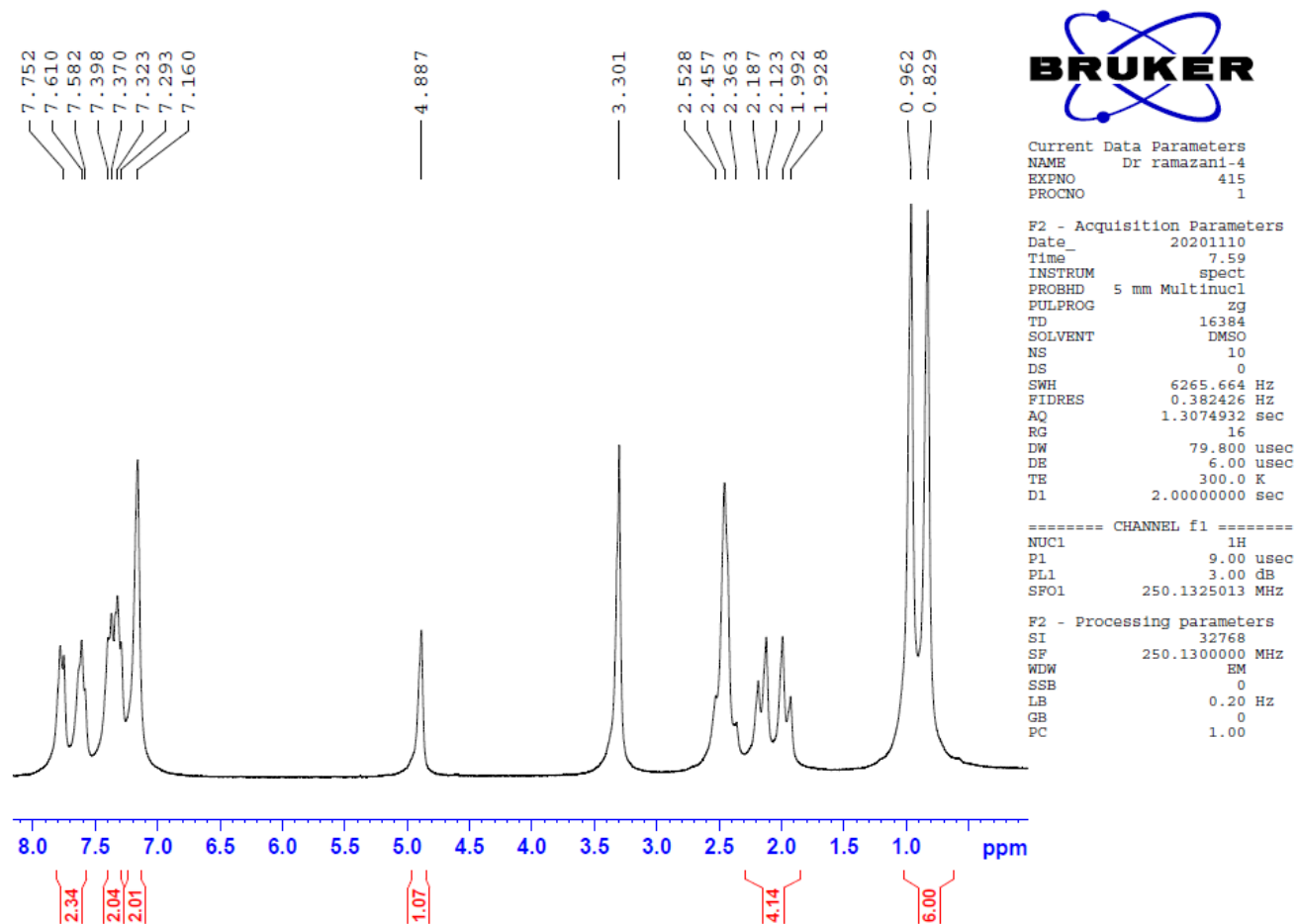
**Figure S4:**  $^{13}\text{C}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(4-cyanophenyl)-5-oxo-5, 6, 7, 8-tetrahydrobenzo[b]pyran (solvent in purity due to in presence of EtOH,  $\delta = 18.9$  and 57.7)



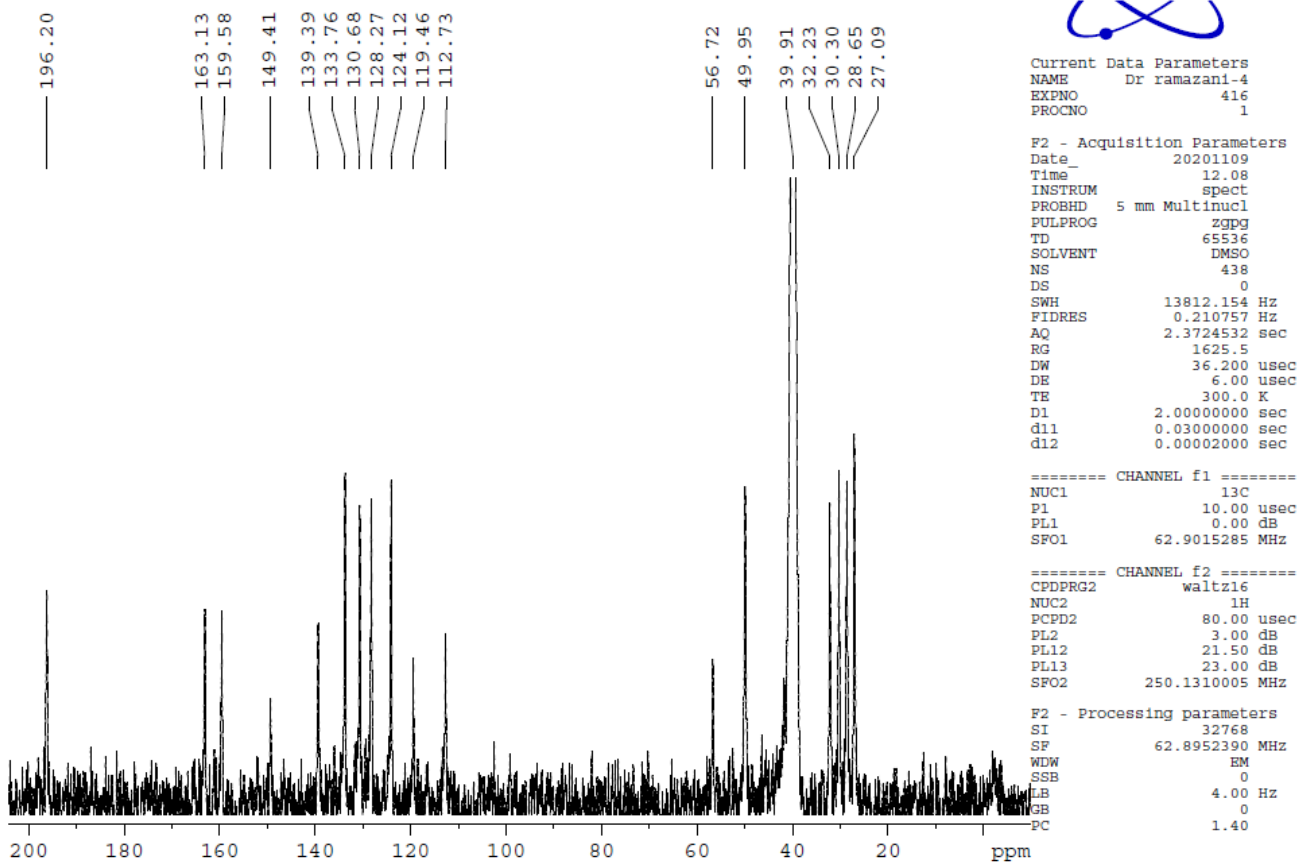
**Figure S5:** FT-IR spectrum of 2-amino-7,7-dimethyl-4-(2-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



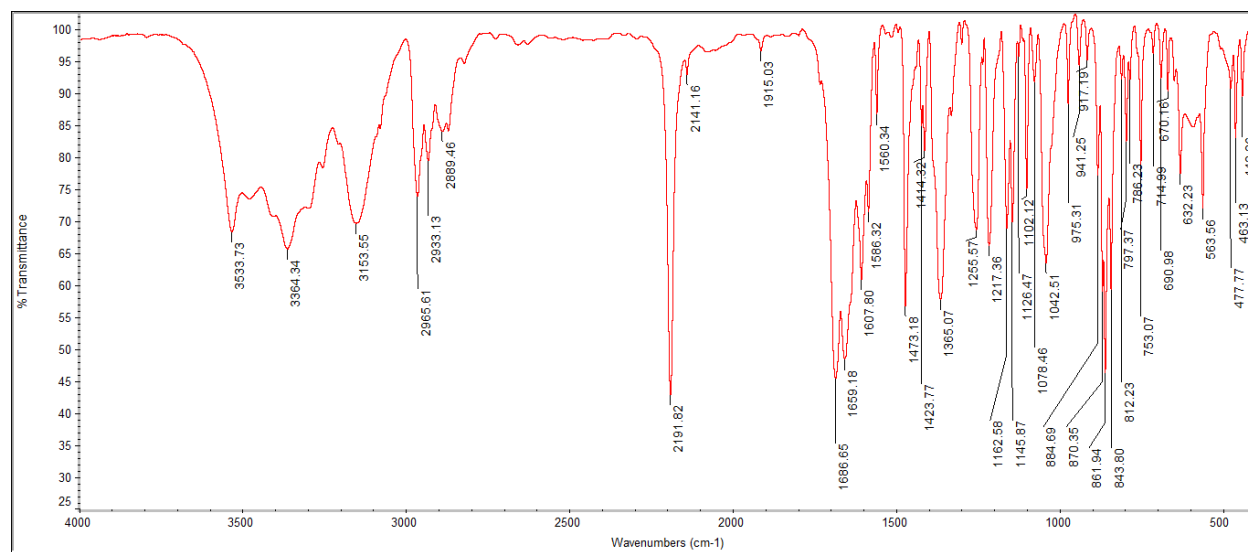
**Figure S6:**  $^1\text{H-NMR}$  spectrum of 2-amino-7,7-dimethyl-4-(2-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



**Figure S7:** The expanded  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(2-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

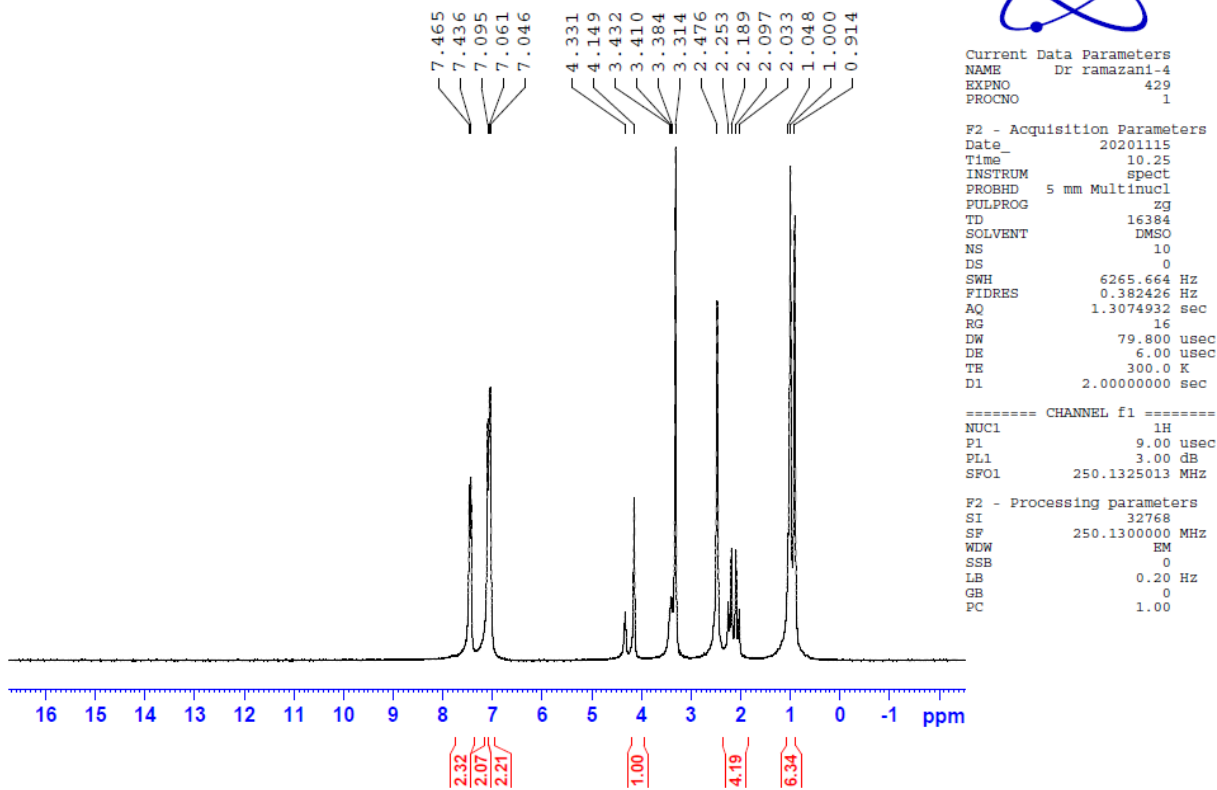


**Figure S8:**  $^{13}\text{C}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(2-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

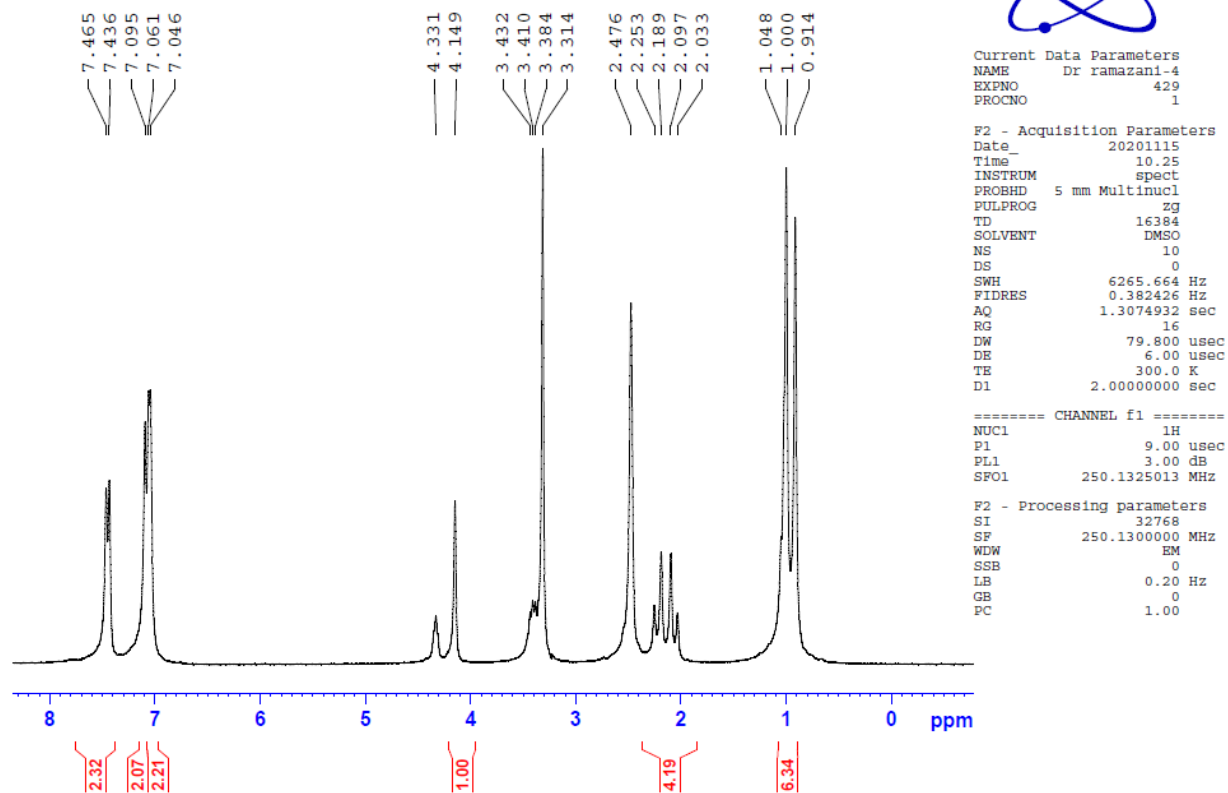


**Figure S9:** FT-IR spectrum of 2-amino-7,7-dimethyl-4-(4-bromophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

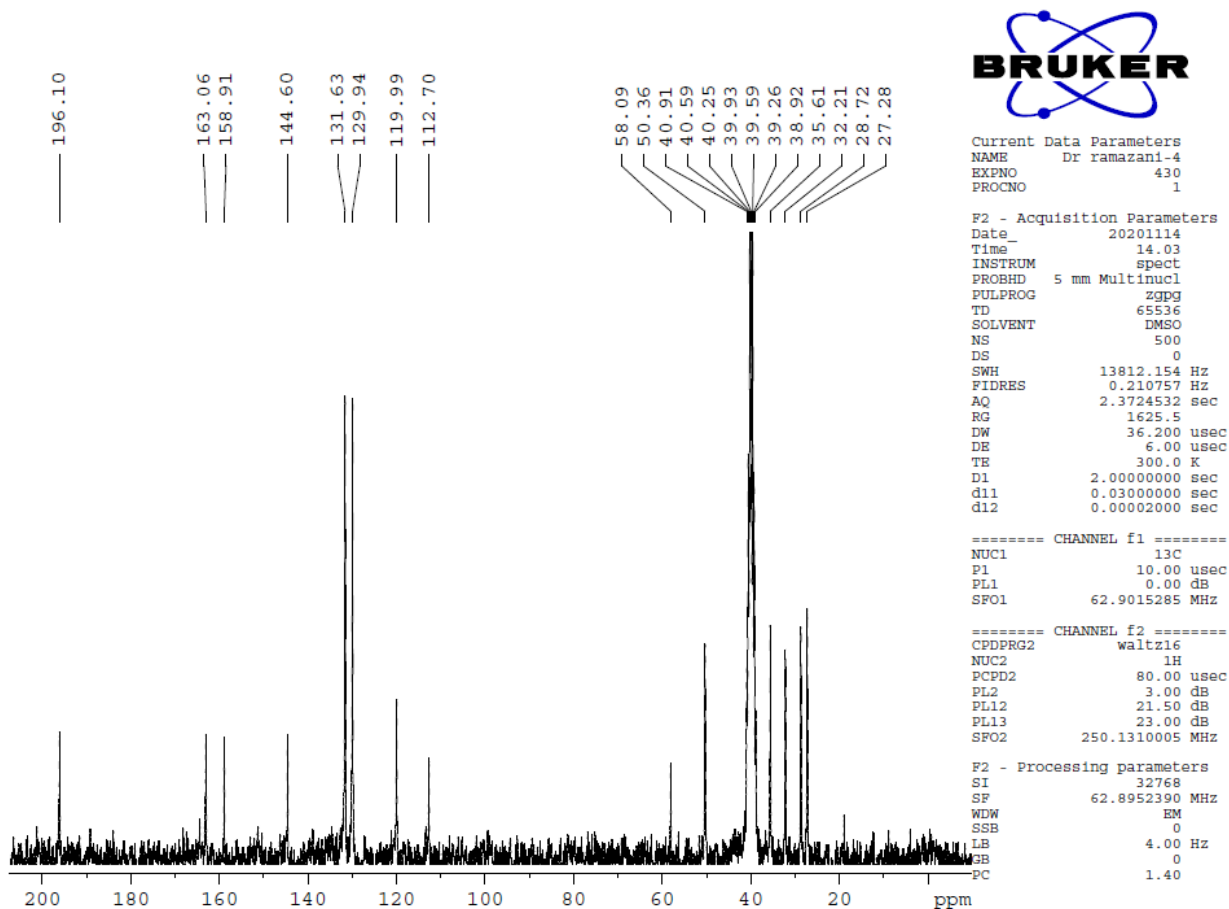




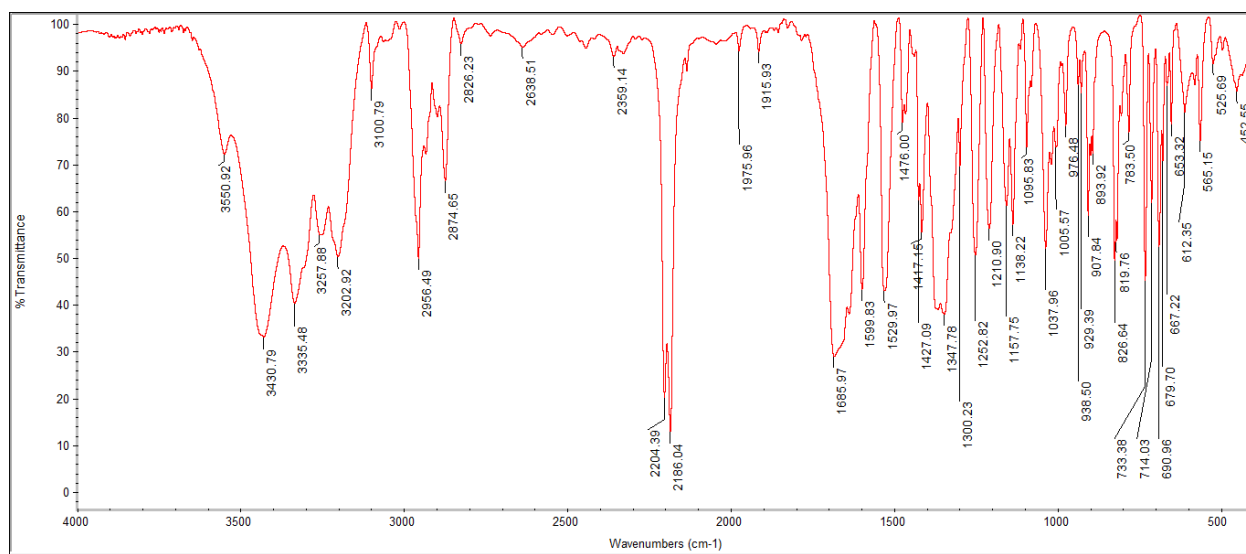
**Figure S10:**  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(4-bromophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



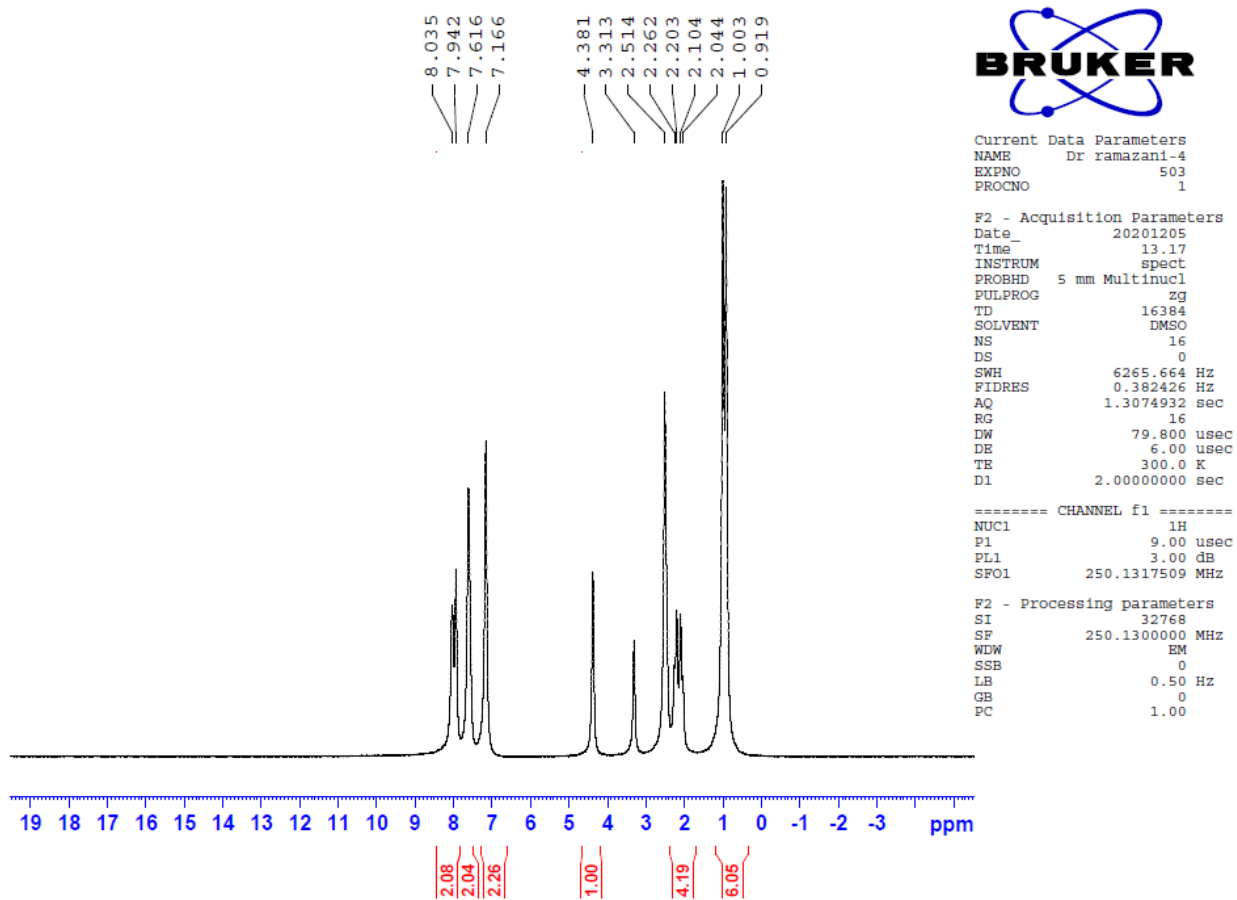
**Figure S11:** The expanded <sup>1</sup>H-NMR spectrum of 2-amino-7,7-dimethyl-4-(4-bromophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



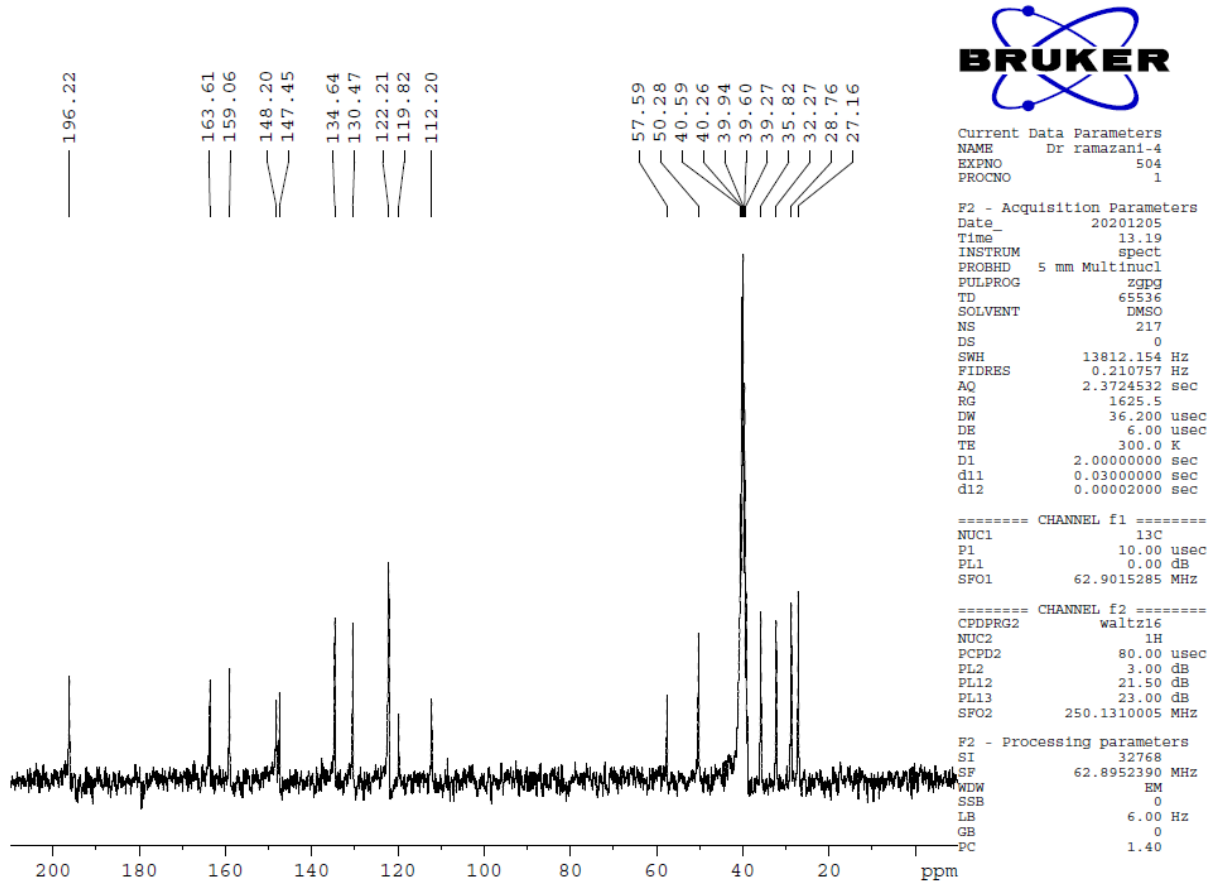
**Figure S12:**  $^{13}\text{C}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(4-bromophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



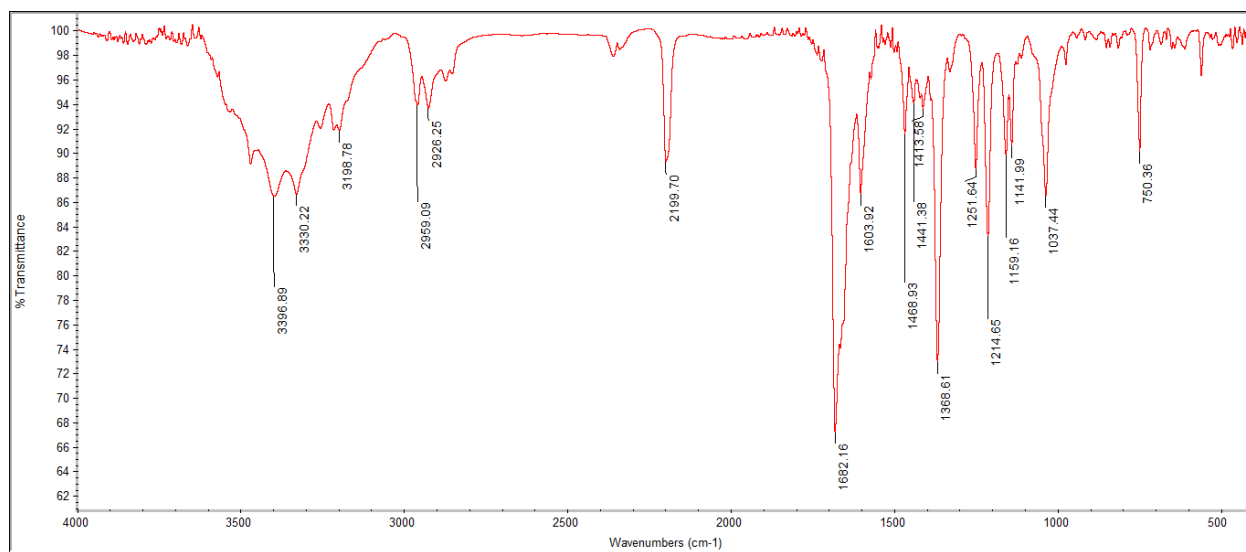
**Figure S13:** FT-IR spectrum of 2-amino-7,7-dimethyl-4-(3-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



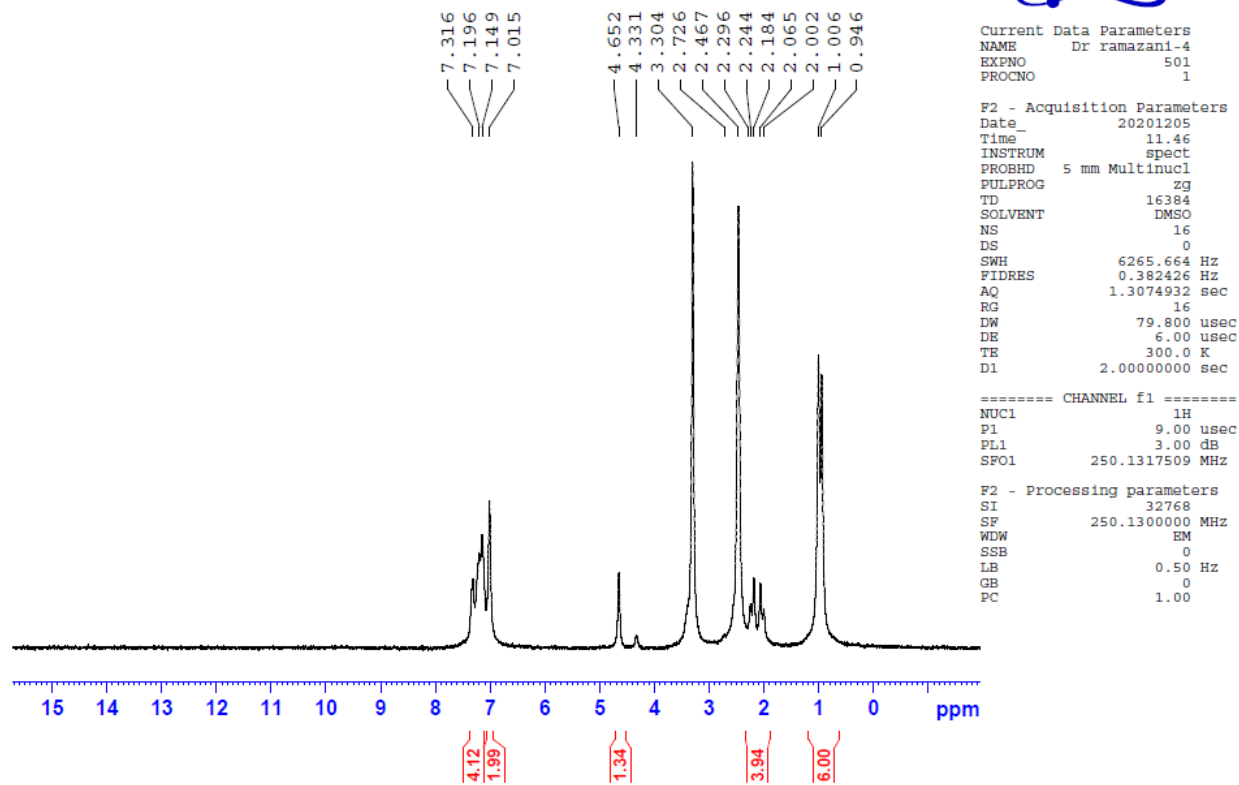
**Figure S14:**  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(3-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



**Figure S15:** <sup>13</sup>C-NMR spectrum of 2-amino-7,7-dimethyl-4-(3-nitrophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

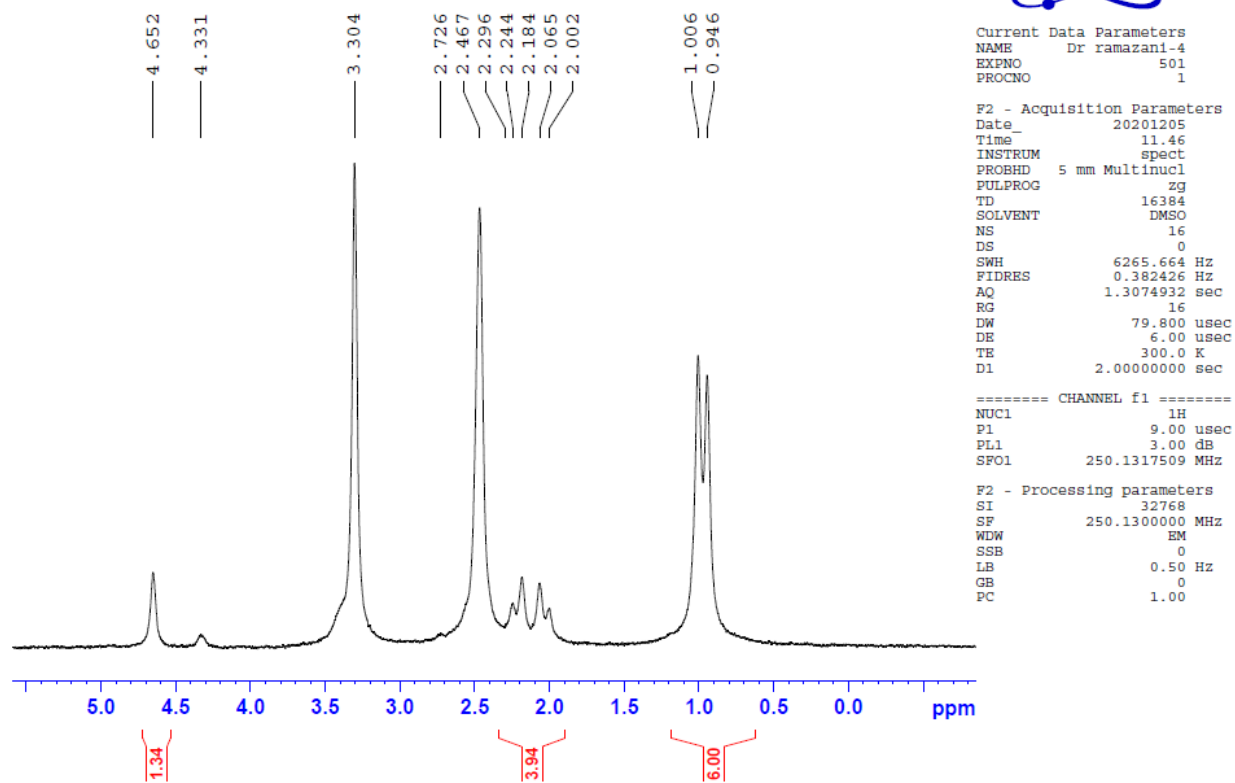


**Figure S16:** FT-IR spectrum of 2-amino-7,7-dimethyl-4-(2-chlorophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

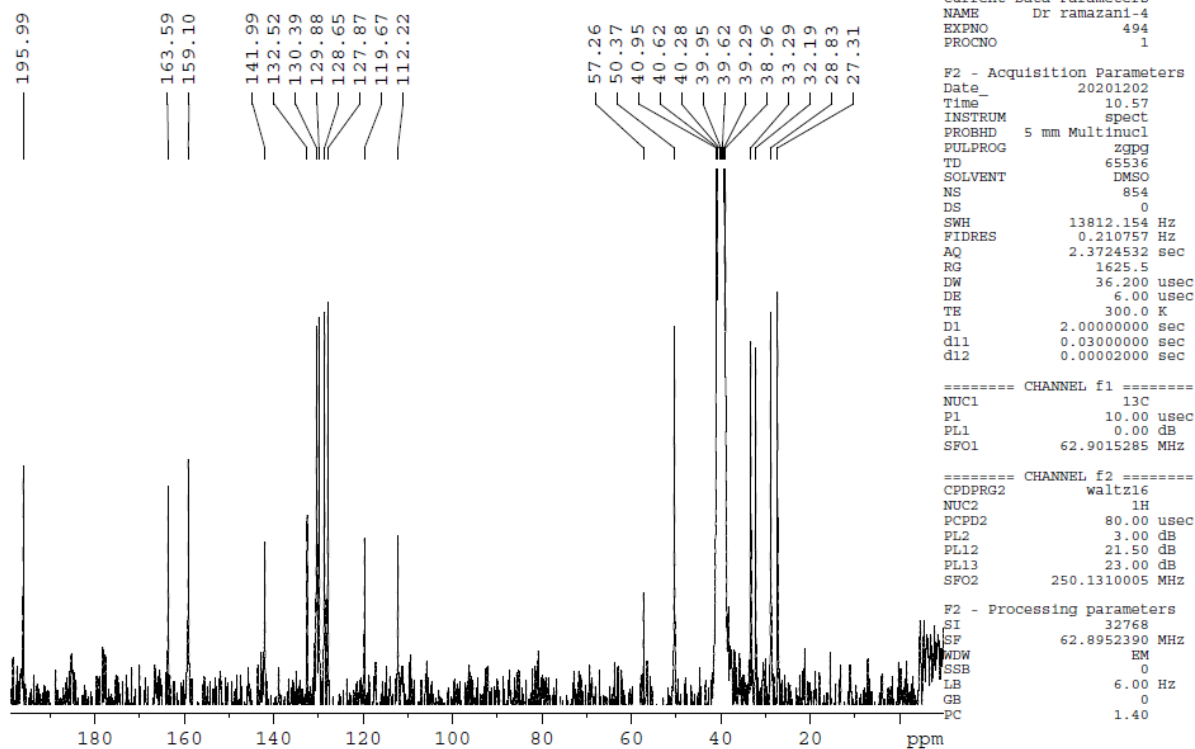


**Figure S17:**  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(2-chlorophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran

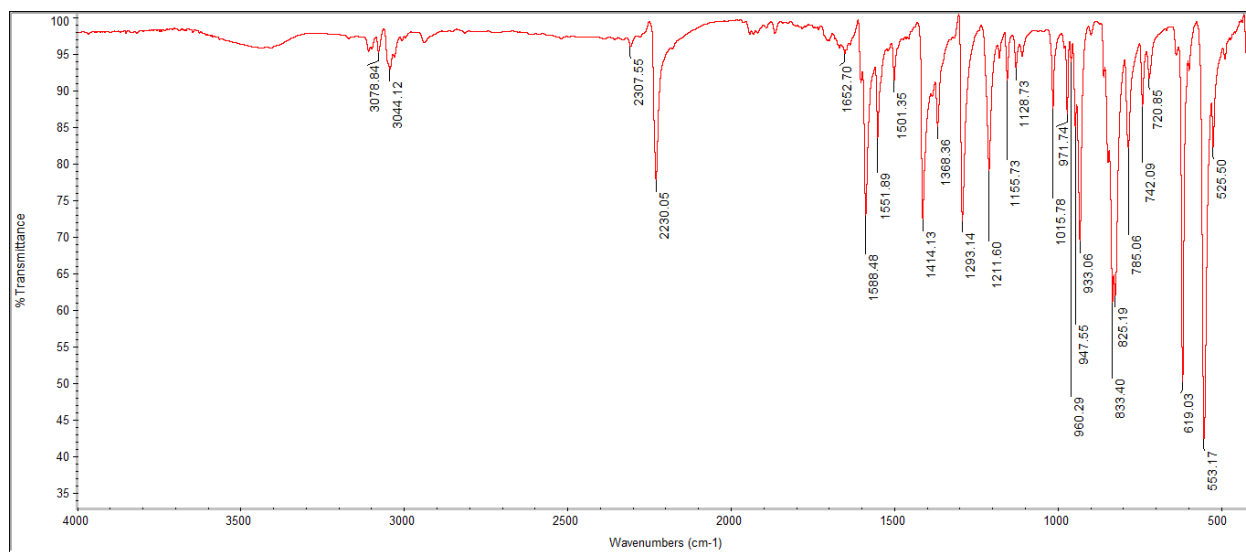




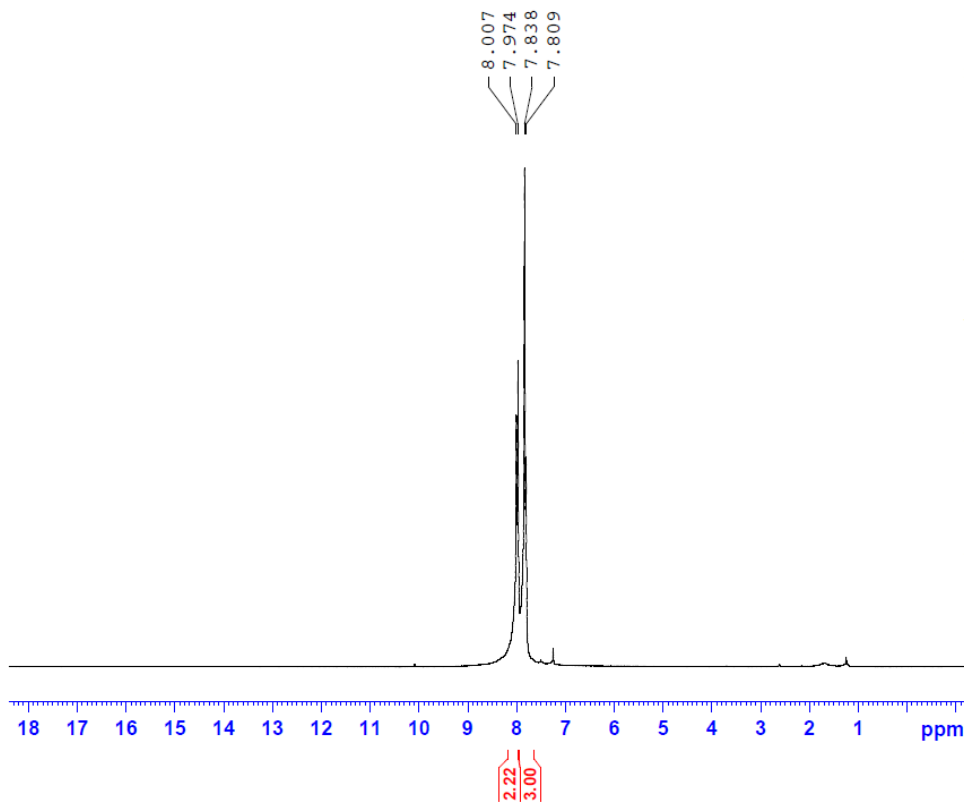
**Figure S18:** The expanded  $^1\text{H}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(2-chlorophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



**Figure S19:**  $^{13}\text{C}$ -NMR spectrum of 2-amino-7,7-dimethyl-4-(2-chlorophenyl)-5-oxo-5,6,7,8-tetrahydrobenzo[b]pyran



**Figure S20:** FT-IR spectrum of 2-(4-cyanobenzylidene)malononitrile



Current Data Parameters  
NAME Dr ramazani-4  
EXPNO 240  
PROCNO 1

F2 - Acquisition Parameters  
Date\_ 20200613  
Time 9.01  
INSTRUM spect  
PROBHD 5 mm Multinucl  
PULPROG zg  
TD 16384  
SOLVENT CDCl3  
NS 16  
DS 0  
SWH 6265.664 Hz  
FIDRES 0.382426 Hz  
AQ 1.3074932 sec  
RG 16  
DW 79.800 usec  
DE 6.00 usec  
TE 300.0 K  
D1 2.0000000 sec

=====  
CHANNEL f1  
NUC1 1H  
P1 9.00 usec  
PL1 3.00 dB  
SFO1 250.1325013 MHz

F2 - Processing parameters  
SI 32768  
SF 250.1300000 MHz  
WDW EM  
SSB 0  
LB 0.50 Hz  
GB 0  
PC 1.00

Figure S21: <sup>1</sup>H-NMR spectrum of 2-(4-cyanobenzylidene)malononitrile

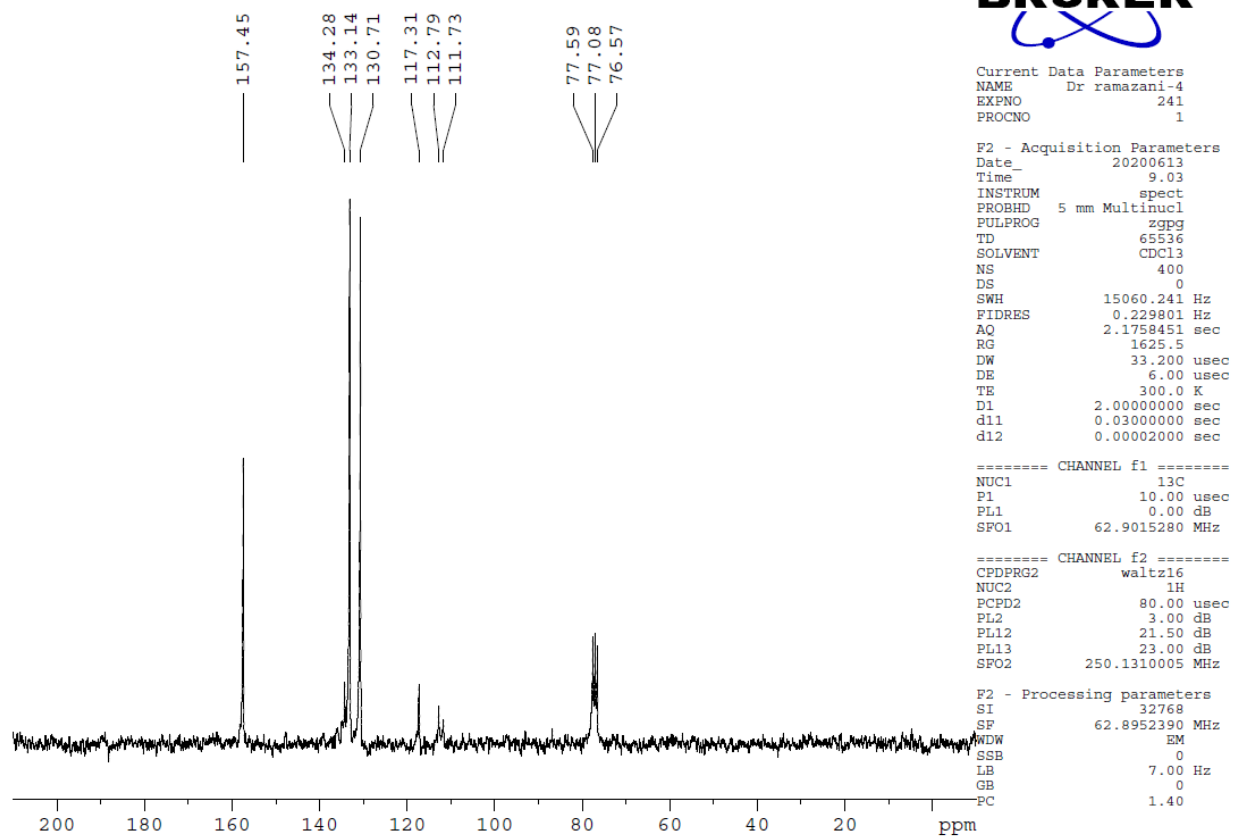
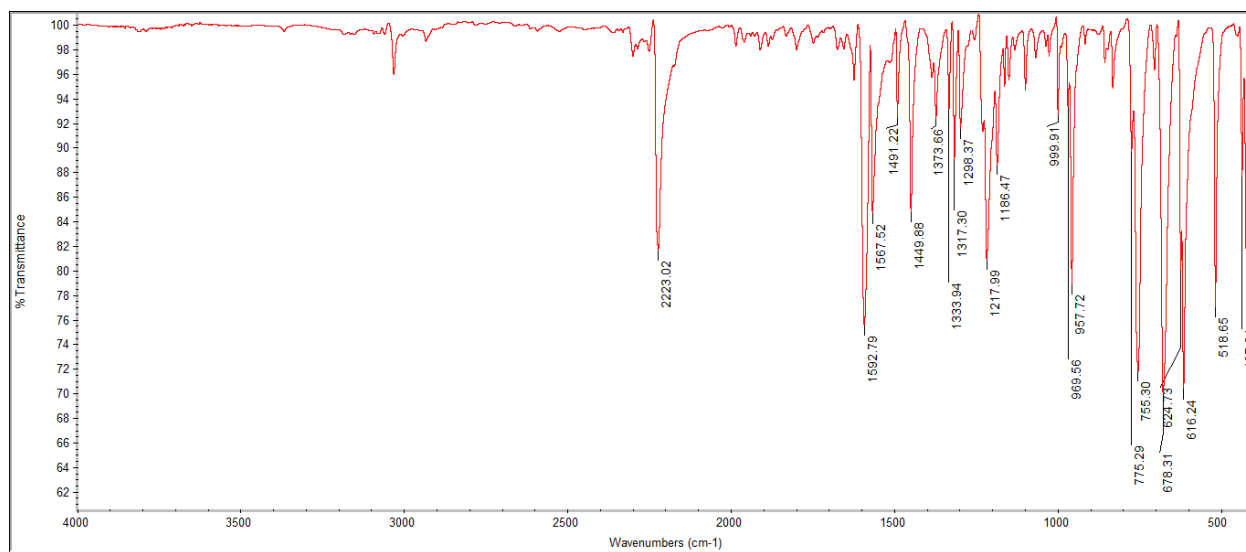
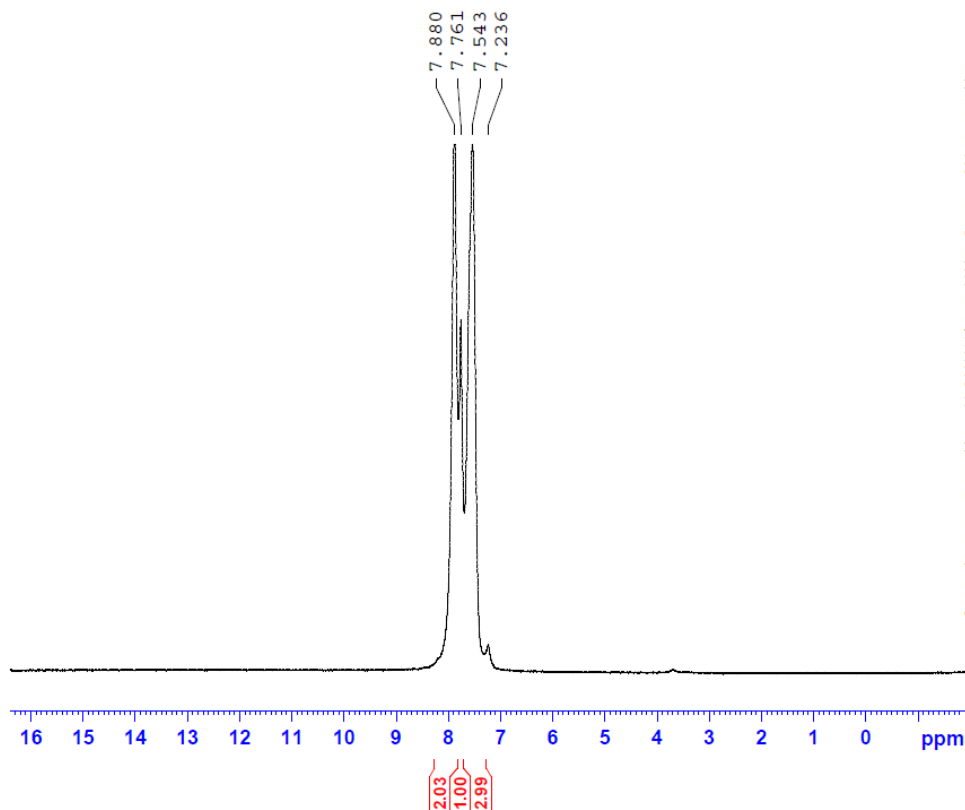


Figure S22:  $^{13}\text{C}$ -NMR spectrum of 2-(4-cyanobenzylidene)malononitrile



**Figure 23:** FT-IR spectrum of 2-benzylidenemalononitrile



Current Data Parameters  
NAME Dr ramazani-4  
EXPNO 210  
PROCNO 1

F2 - Acquisition Parameters  
Date\_ 20200609  
Time 8.16  
INSTRUM spect  
PROBHD 5 mm Multinucl  
PULPROG zg  
TD 16384  
SOLVENT CDCl3  
NS 10  
DS 0  
SWH 6265.664 Hz  
FIDRES 0.382426 Hz  
AQ 1.3074932 sec  
RG 16  
DW 79.800 usec  
DE 6.00 usec  
TE 300.0 K  
D1 2.0000000 sec

===== CHANNEL f1 =====  
NUC1 1H  
P1 9.00 usec  
PL1 3.00 dB  
SFO1 250.1325013 MHz

F2 - Processing parameters  
SI 32768  
SF 250.1300000 MHz  
WDW EM  
SSB 0  
LB 0.50 Hz  
GB 0  
PC 1.00

Figure 24:  $^1\text{H}$ -NMR spectrum of 2-benzylidenemalononitrile

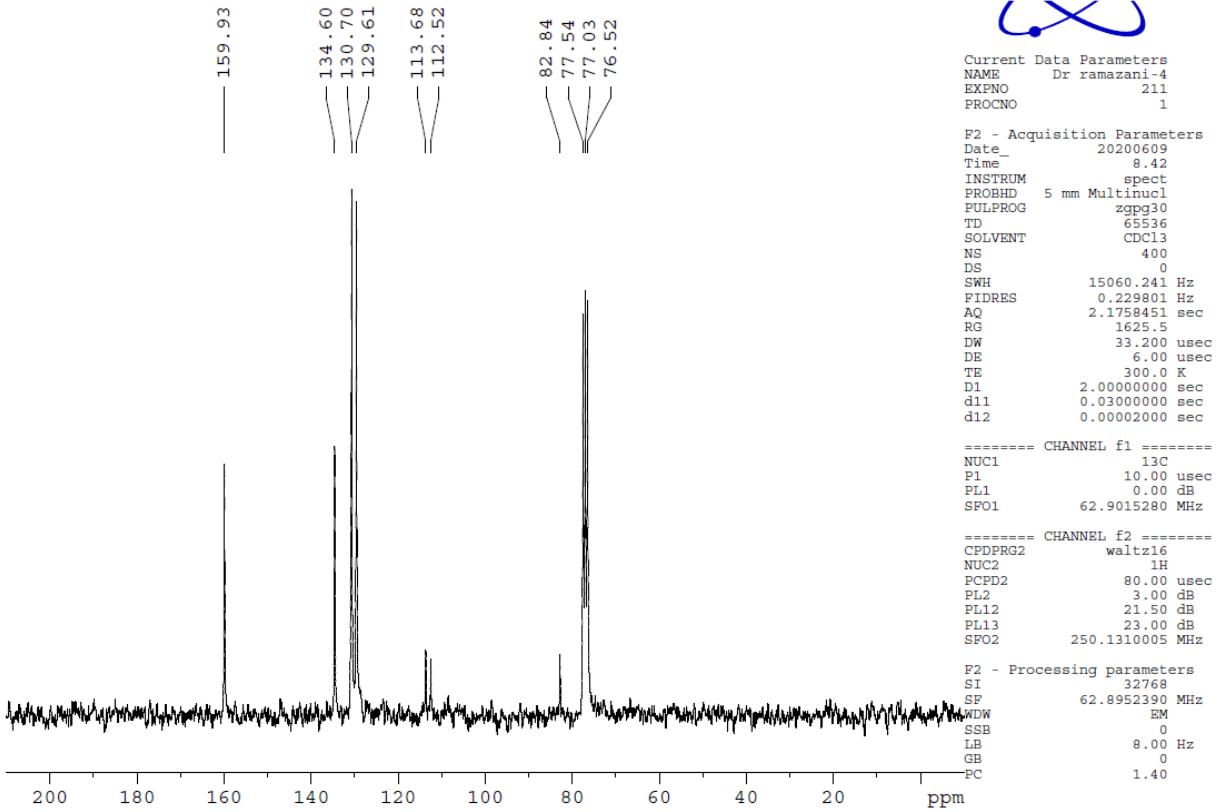
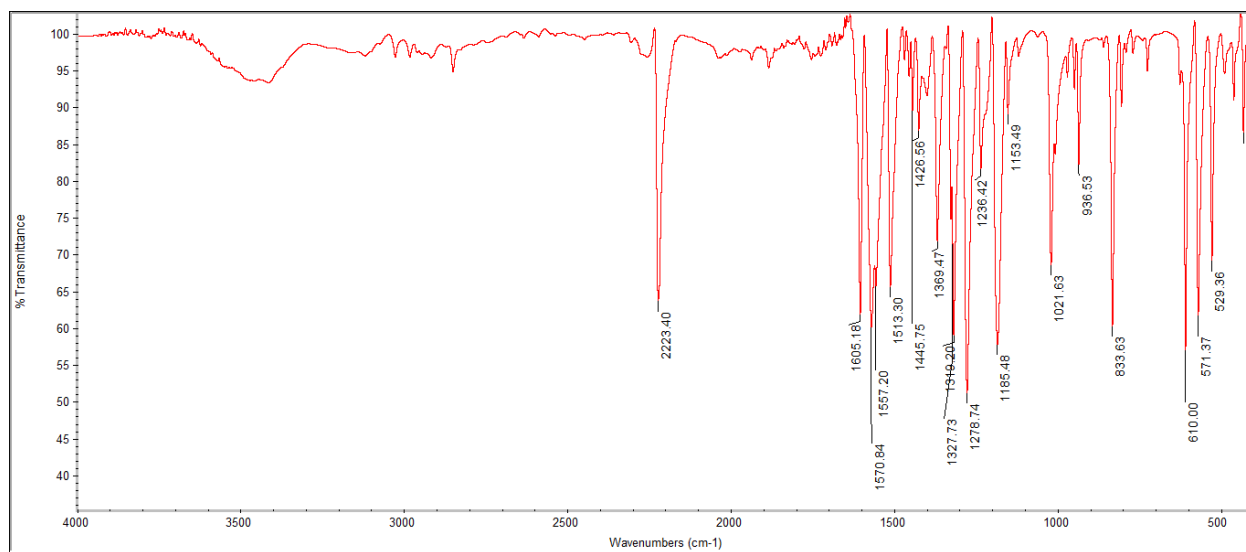


Figure 25:  $^{13}\text{C}$ -NMR spectrum of 2-benzylidenemalononitrile





**Figure 26:** FT-IR spectrum of 2-(4-methoxybenzylidene)malononitrile

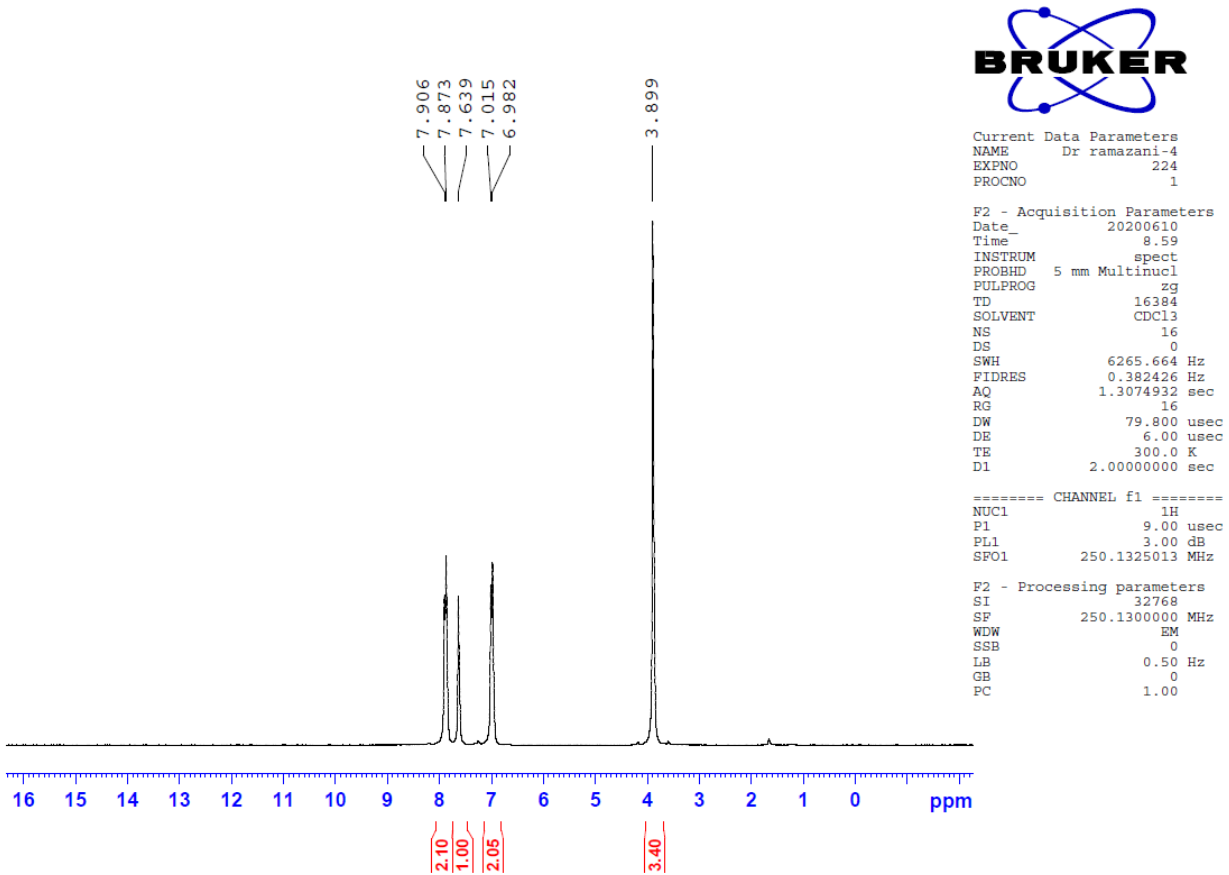
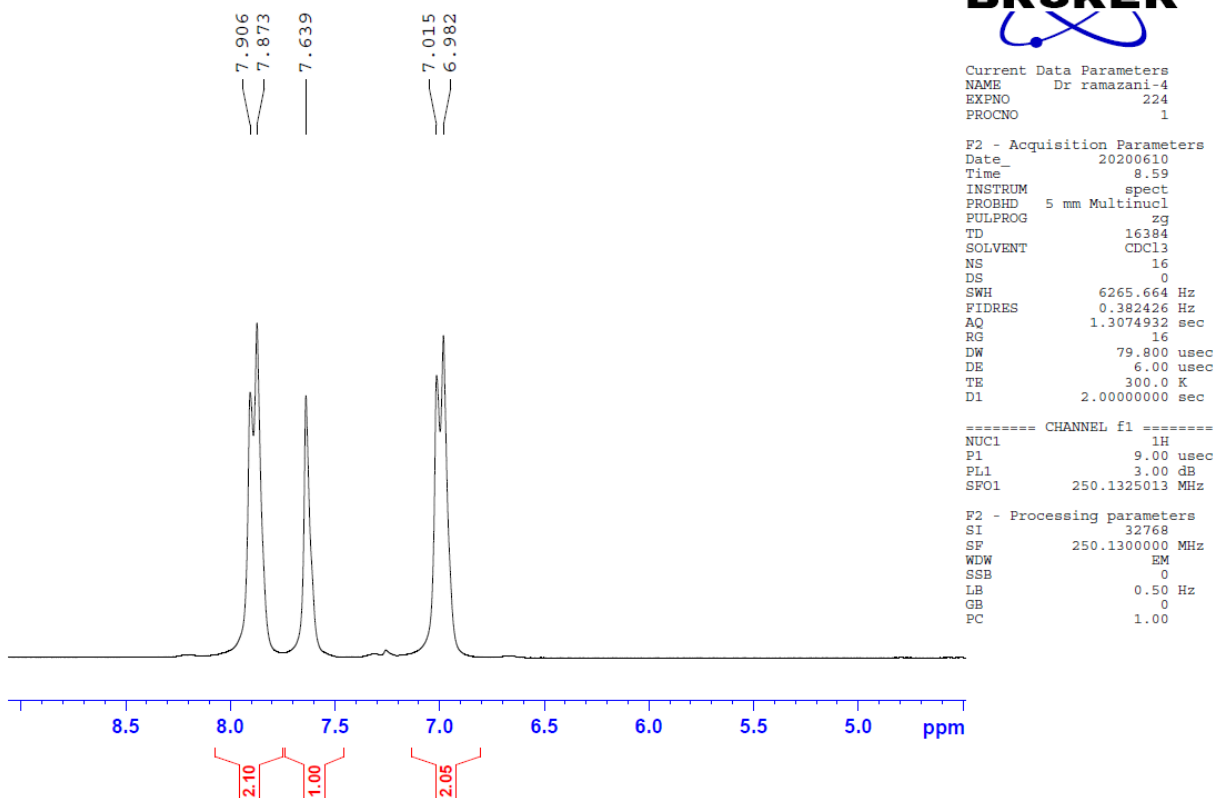


Figure 27: <sup>1</sup>H-NMR spectrum of 2-(4-methoxybenzylidene)malononitrile



**Figure 28:** The expanded  $^1\text{H}$ -NMR spectrum of 2-(4-methoxybenzylidene)malononitrile

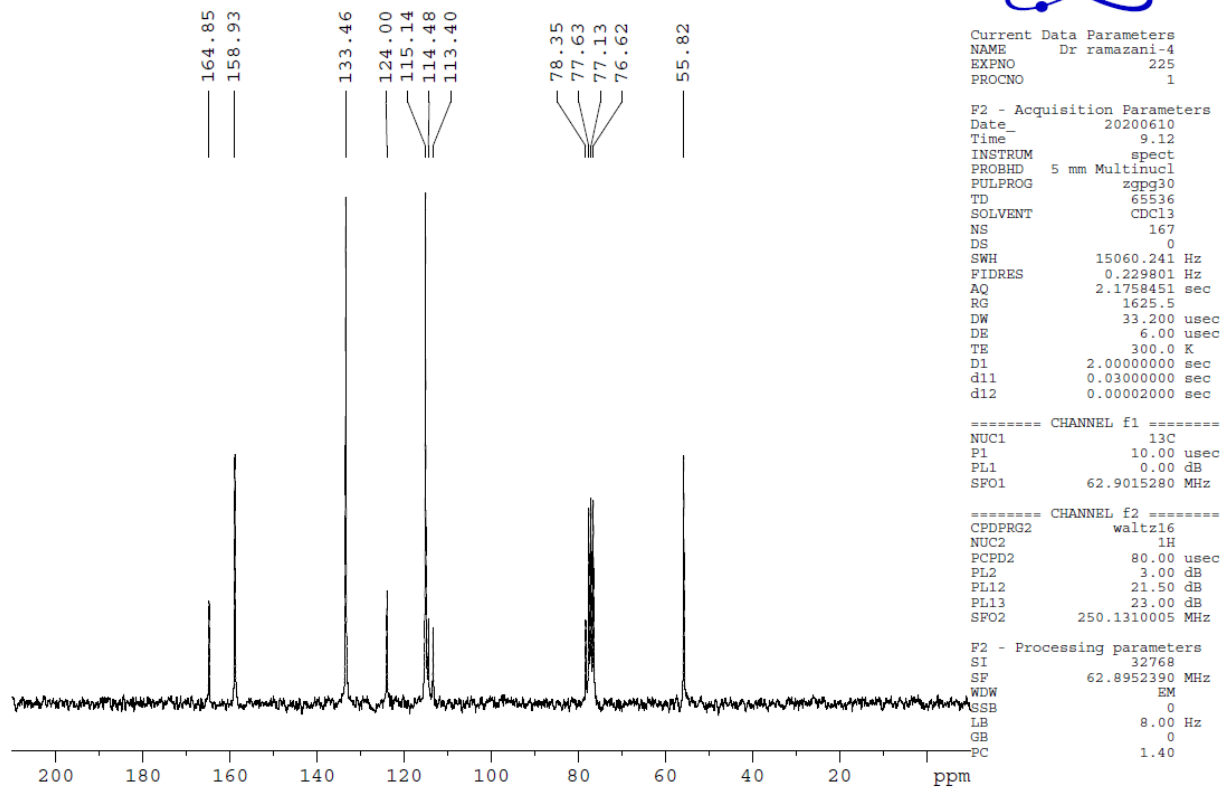
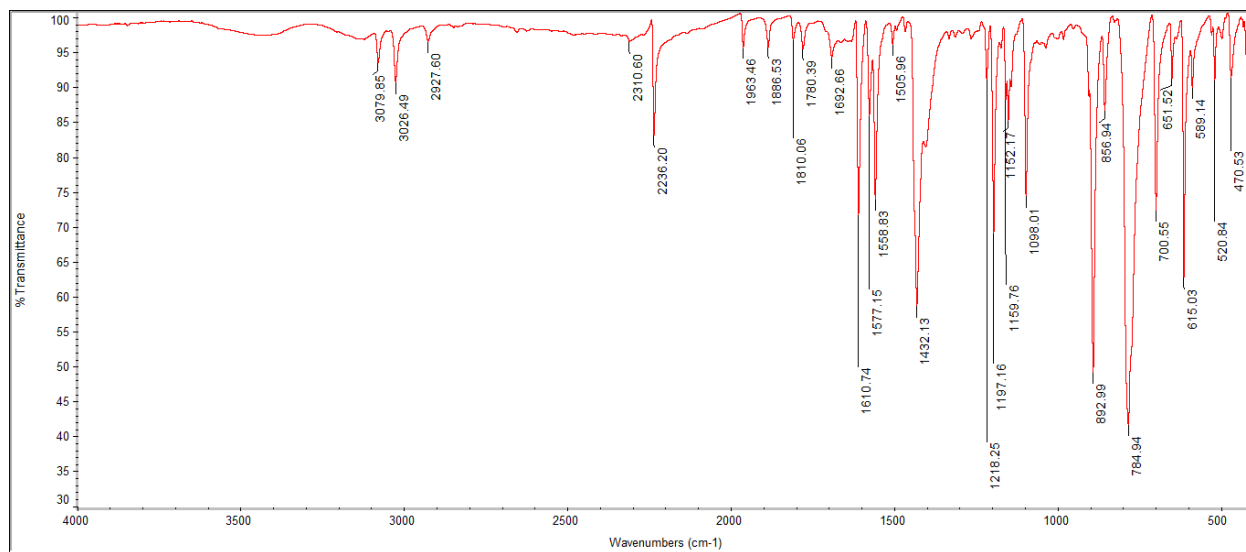
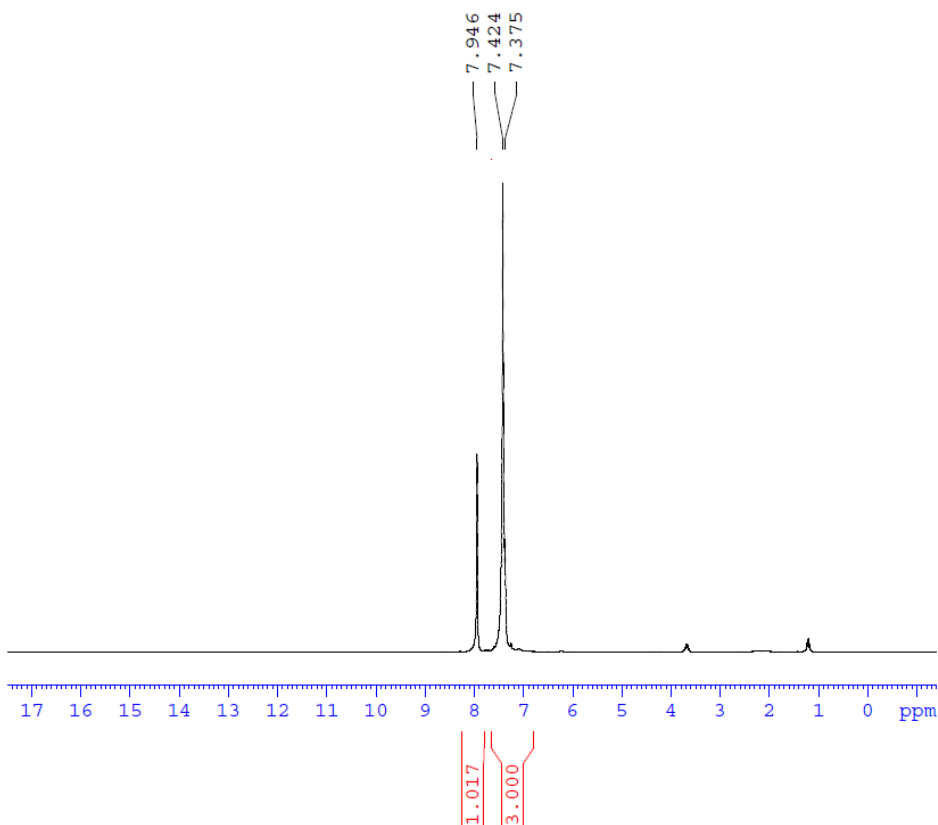


Figure 29:  $^{13}\text{C}$ -NMR spectrum of 2-(4-methoxybenzylidene)malononitrile



**Figure 30:** FT-IR spectrum of 2-(2,6-dichlorobenzylidene)malononitrile



Current Data Parameters  
NAME Dr ramazani-4  
EXPNO 254  
PROCNO 1

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Date\_ 20200615  
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PULPROG zg  
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SOLVENT CDCl3  
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DS 0  
SWH 6265.664 Hz  
FIDRES 0.382426 Hz  
AQ 1.3074932 sec  
RG 16  
DW 79.800 usec  
DE 6.00 usec  
TE 300.0 K  
D1 2.00000000 sec

===== CHANNEL f1 =====  
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P1 9.00 usec  
PL1 2.00 dB  
SFO1 250.1325013 MHz

F2 - Processing parameters  
SI 32768  
SF 250.1300000 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0  
PC 1.00

Figure 31: <sup>1</sup>H-NMR spectrum of 2-(2,6-dichlorobenzylidene)malononitrile

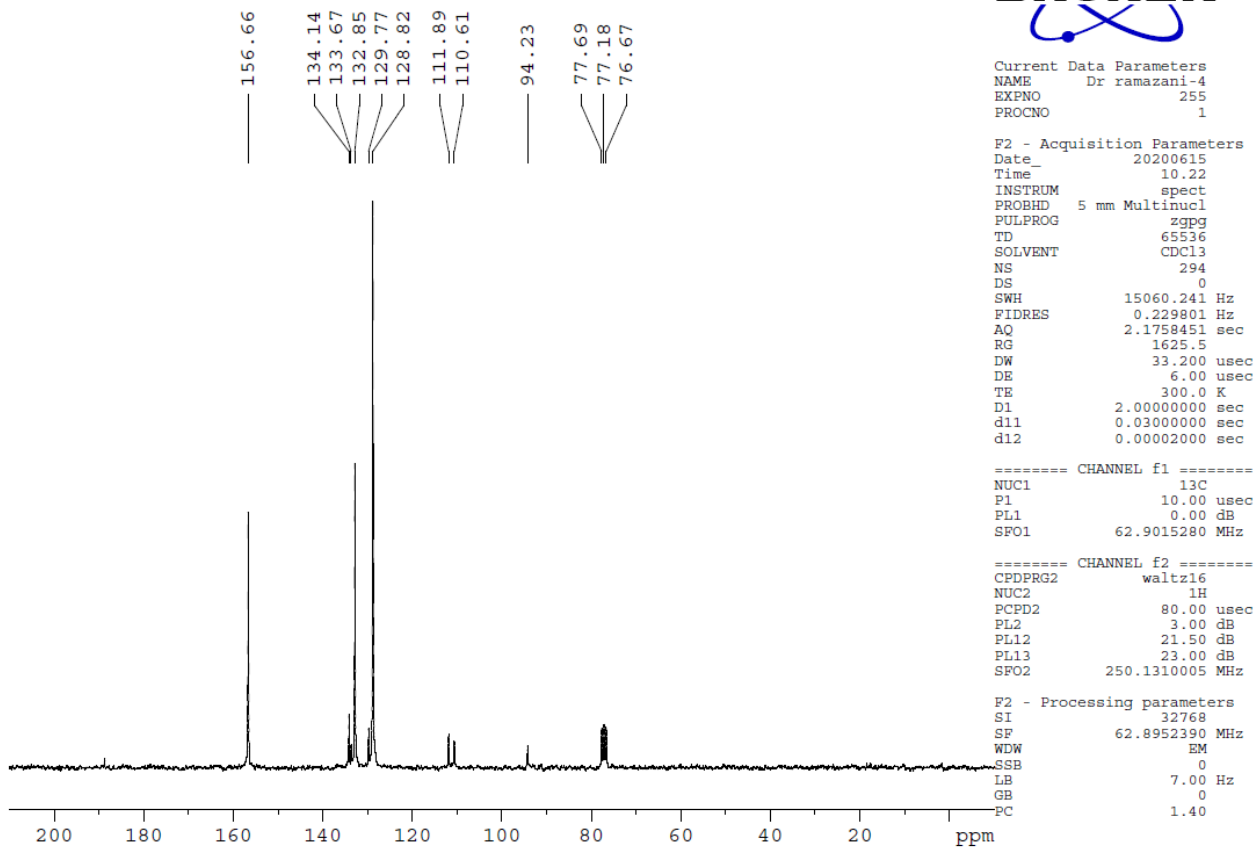
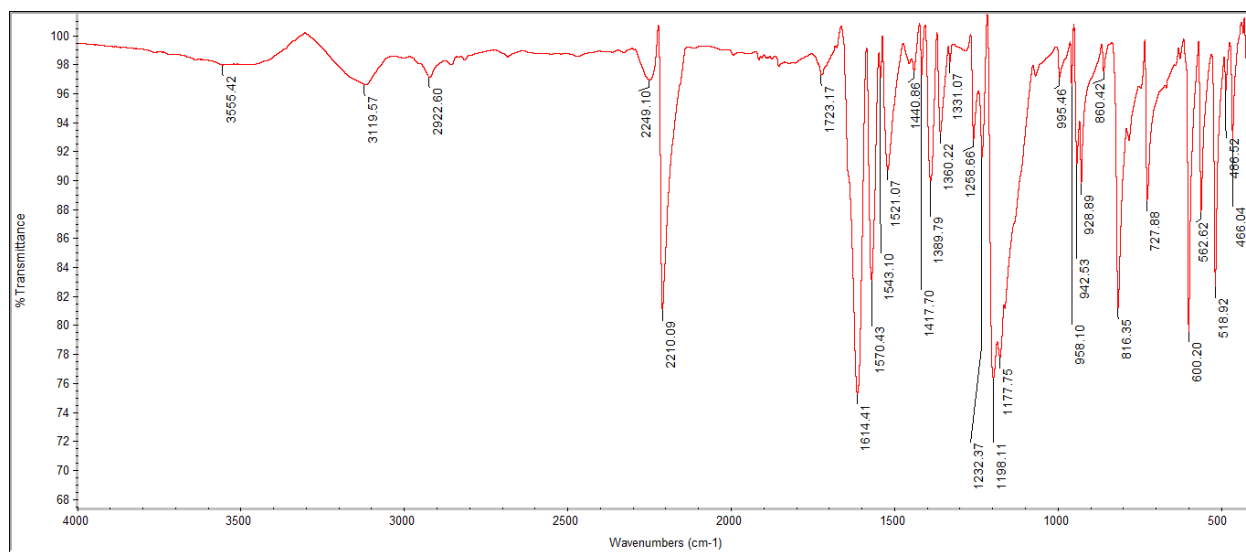
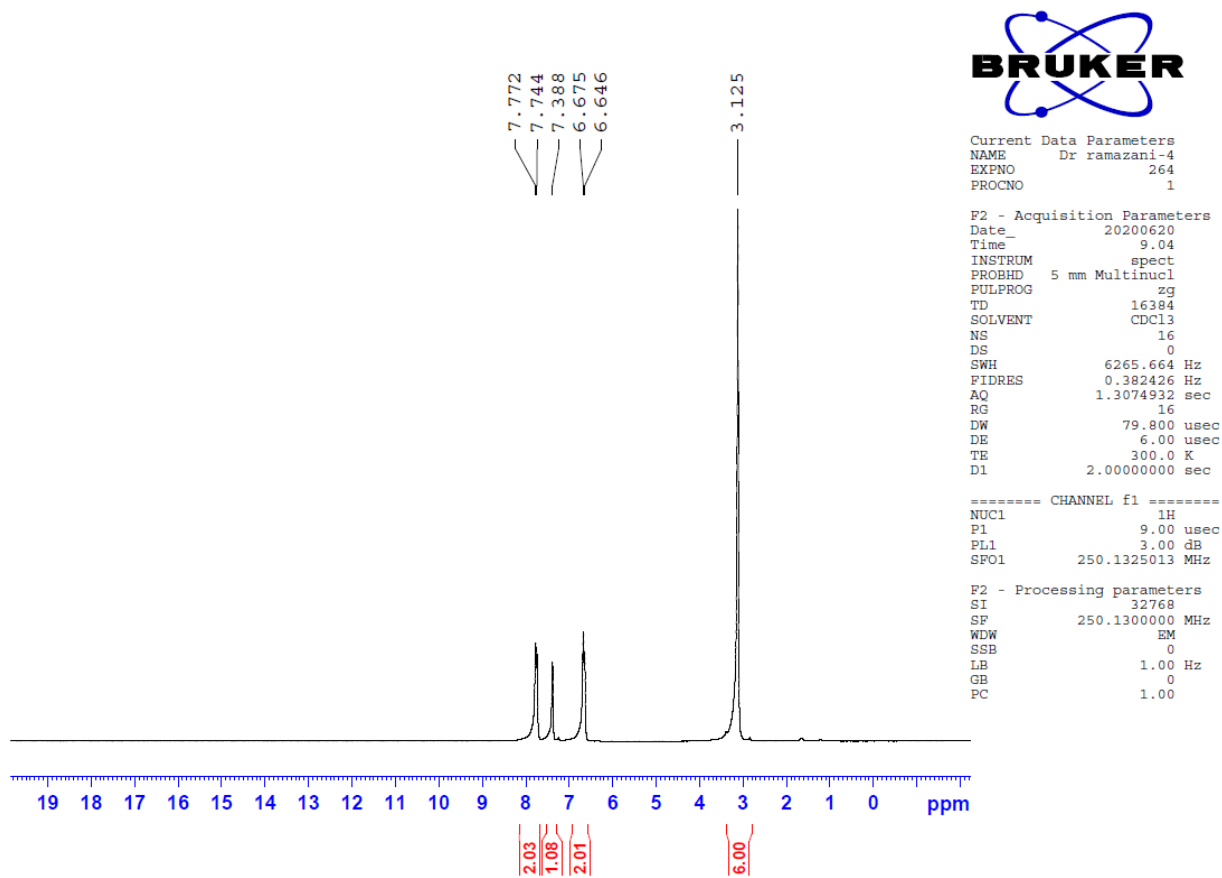


Figure 32:  $^{13}\text{C}$ -NMR spectrum of 2-(2,6-dichlorobenzylidene)malononitrile

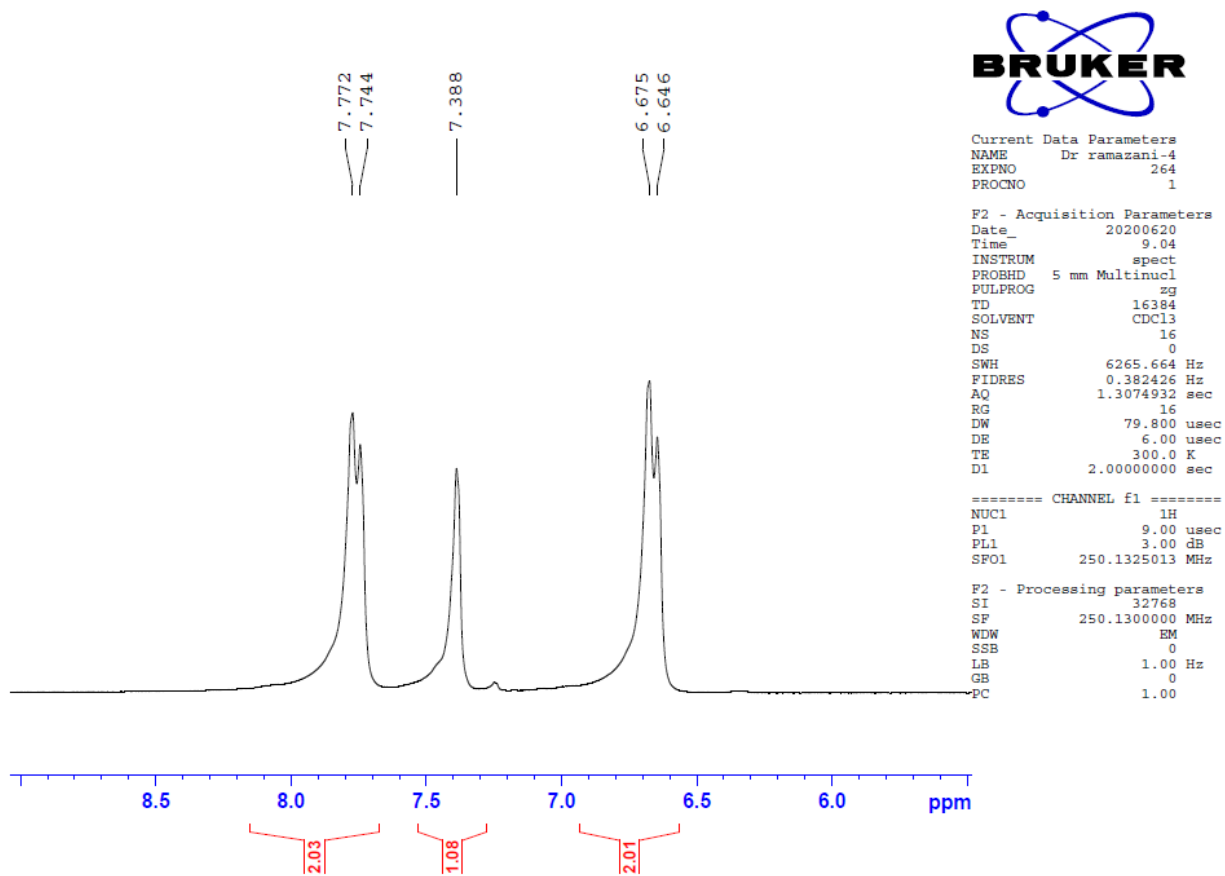


**Figure 33:** FT-IR spectrum of 2-(4-(dimethylamino)benzylidene)malononitrile





**Figure 34:**  $^1\text{H}$ -NMR spectrum of 2-(4-(dimethylamino)benzylidene)malononitrile



**Figure 35:** The expanded  $^1\text{H}$ -NMR spectrum of 2-(4-(dimethylamino)benzylidene)malononitrile

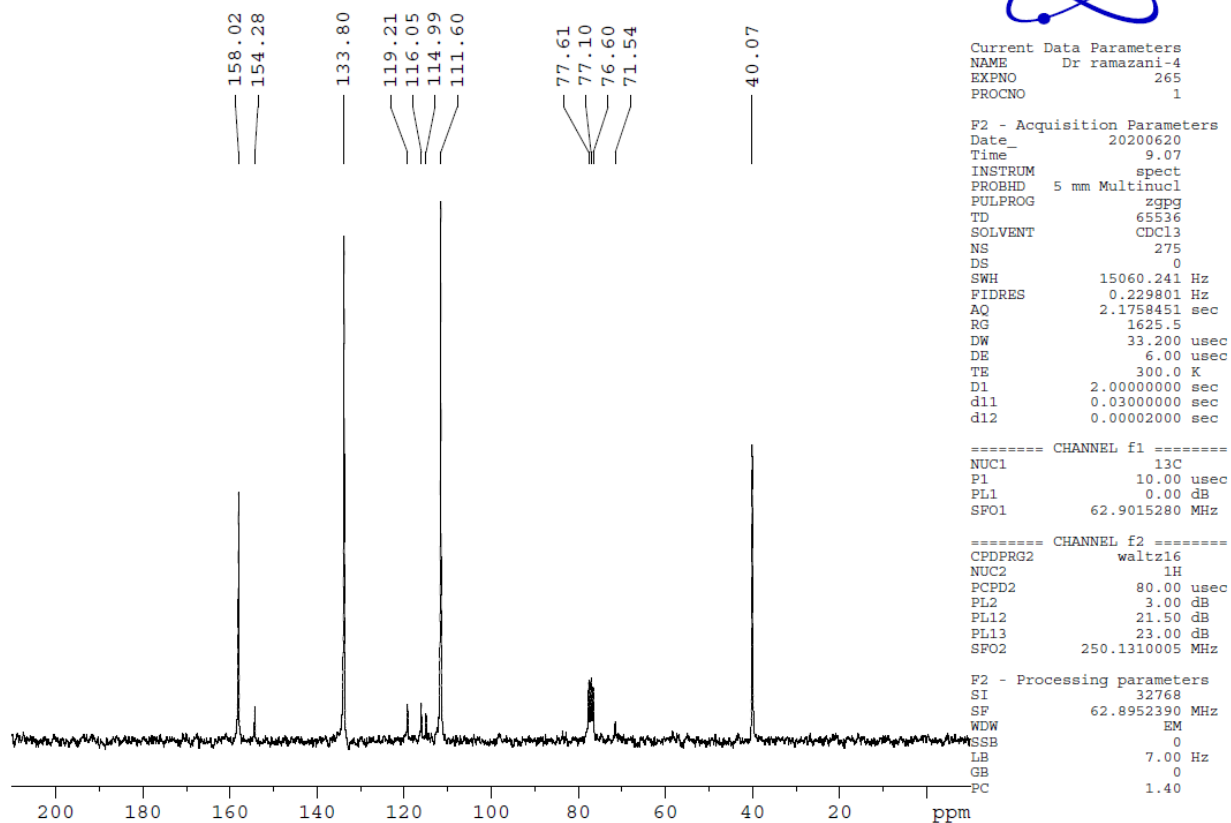


Figure 36:  $^{13}\text{C}$ -NMR spectrum of 2-(4-(dimethylamino)benzylidene)malononitrile