

## Supporting Information

### Graphitic Carbon Nitride Nanosheet/FeWO<sub>4</sub> Nanoparticle Composite for Tandem Photooxidation/Knoevenagel Condensation

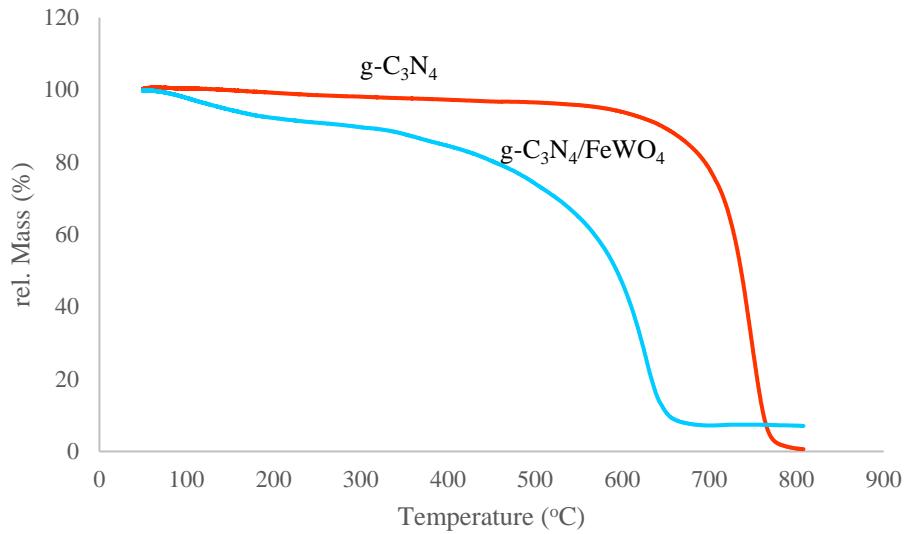
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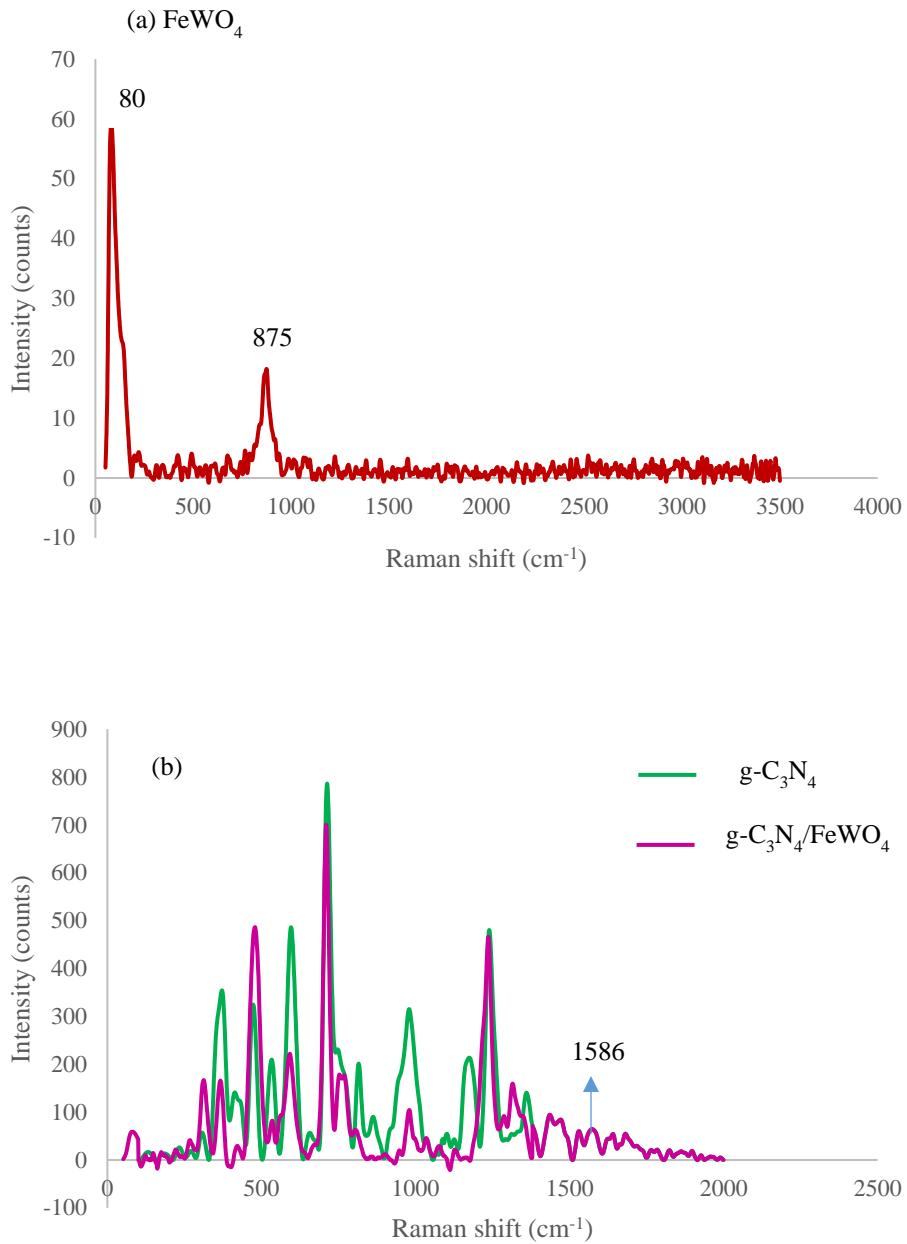
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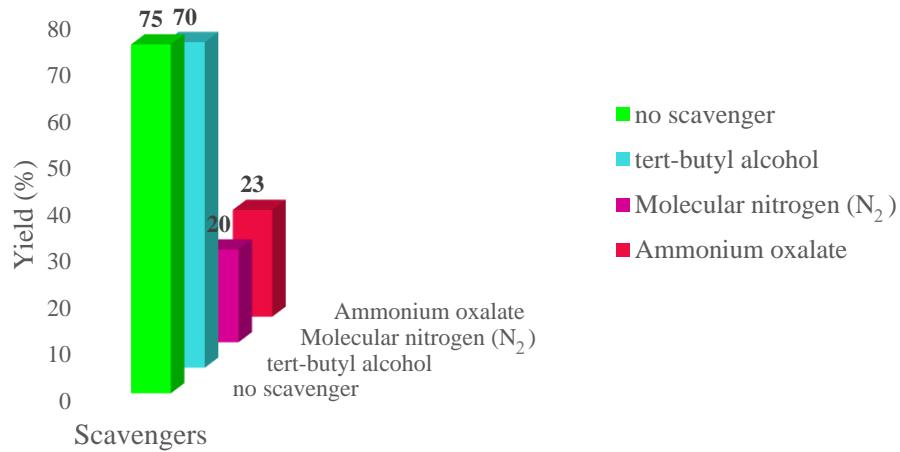
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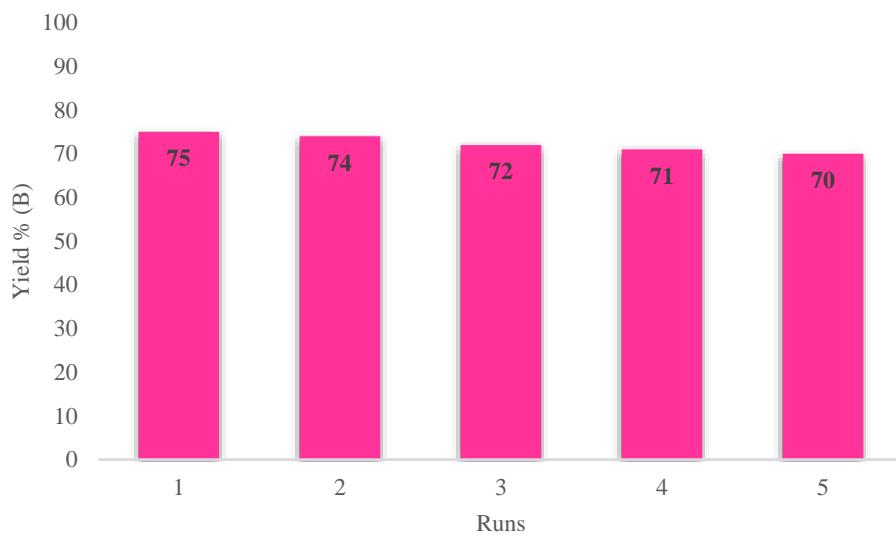
**Figure S1.** Thermogravimetric analysis of g-C<sub>3</sub>N<sub>4</sub> and g-C<sub>3</sub>N<sub>4</sub>/FeWO<sub>4</sub>.



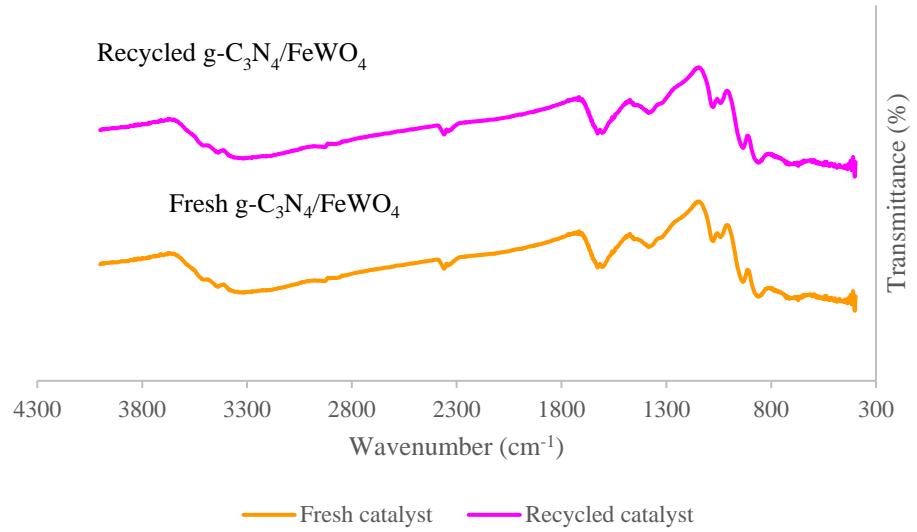
**Figure S2.** Raman spectrum of  $\text{FeWO}_4$  (a),  $\text{g-C}_3\text{N}_4$  and  $\text{g-C}_3\text{N}_4/\text{FeWO}_4$  (b).



**Figure S3.** Trapping experiment of active types of oxidants during the photo-oxidation/Knoevenagel condensation.



**Figure S4.** Recyclability of the  $\text{g-C}_3\text{N}_4/\text{FeWO}_4$  in tandem one-pot photo-oxidation/ Knoevenagel condensation of alcohols.



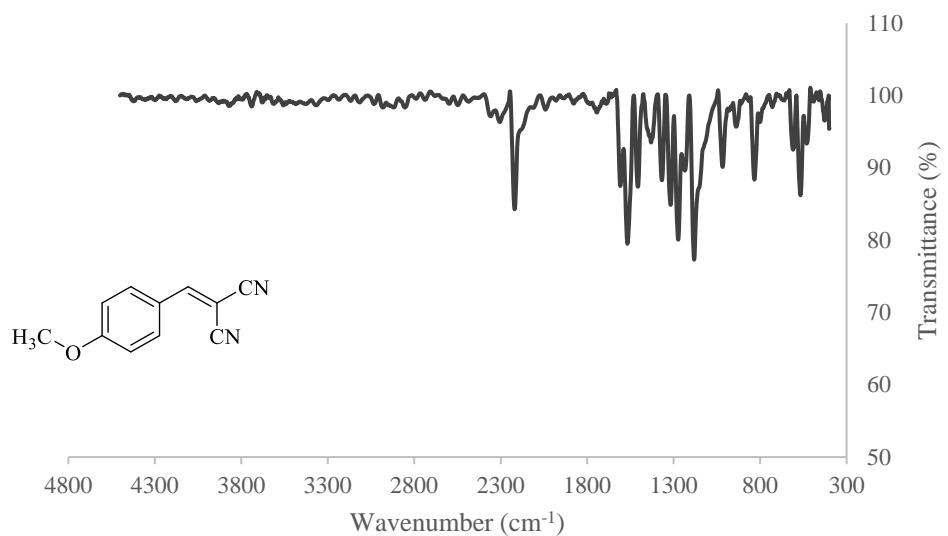
**Figure S5.** FT-IR spectrum of fresh and recycled g-C<sub>3</sub>N<sub>4</sub>/FeWO<sub>4</sub>.

**Table S1.** Optimization of the reaction conditions for the one-pot photo-oxidation/ Knoevenagel condensation catalyzed by g-C<sub>3</sub>N<sub>4</sub>/FeWO<sub>4</sub>.<sup>a</sup>

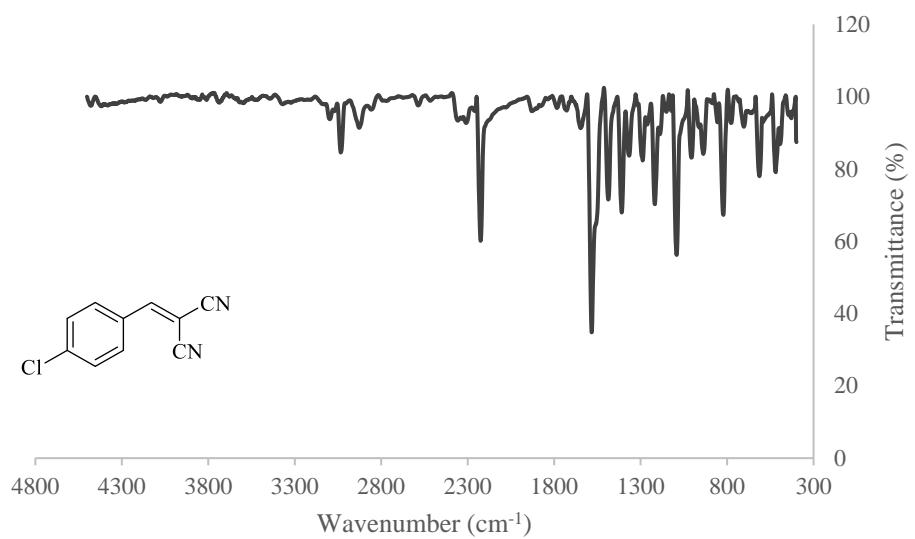
The reaction scheme illustrates the two-step synthesis of product B. In the first step, benzyl alcohol reacts with a catalyst (g-C<sub>3</sub>N<sub>4</sub>/FeWO<sub>4</sub>) in CH<sub>3</sub>CN at room temperature (r.t.) for 10.0 h (Time A) to form intermediate A (benzyl aldehyde). In the second step, intermediate A reacts with malononitrile in CH<sub>3</sub>CN at r.t. for 4.0 h (Time B) to form product B (benzyl malononitrile).

Entry	Catalyst	Amount of catalyst (mg)	Temp. (°C)	Solvent	hν	Condition	Conversion of benzyl alcohol (%)	Yield <sup>b</sup> (B) %
1	-	-	r.t.	CH <sub>3</sub> CN	-	Air	-	-
2	-	-	r.t.	CH <sub>3</sub> CN	+	Air	5	-
3	g-C <sub>3</sub> N <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	-	Air	-	-
4	g-C <sub>3</sub> N <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	Air	35	20
5	g-C <sub>3</sub> N <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub>	45	30
6	g-C <sub>3</sub> N <sub>4</sub>	40	80	CH <sub>3</sub> CN	+	O <sub>2</sub>	30	23
7	FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub>	10	-
8	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	-	Air	-	-
9	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	Air	65	50
10	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub>	84	75
11	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	N <sub>2</sub>	30	20
12	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	80	CH <sub>3</sub> CN	+	O <sub>2</sub>	69	60
13	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub> , H <sub>2</sub> O <sub>2</sub>	35	20
14	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	-	O <sub>2</sub> , H <sub>2</sub> O <sub>2</sub>	10	-
15	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	-	O <sub>2</sub> , TBHP	12	-
16	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub> , TBHP	15	-
17	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	H <sub>2</sub> O	+	O <sub>2</sub>	40	35
18	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	40	r.t.	Toluene	+	O <sub>2</sub>	15	Trace
19	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	30	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub>	75	63
20	g-C <sub>3</sub> N <sub>4</sub> /FeWO <sub>4</sub>	50	r.t.	CH <sub>3</sub> CN	+	O <sub>2</sub>	80	70

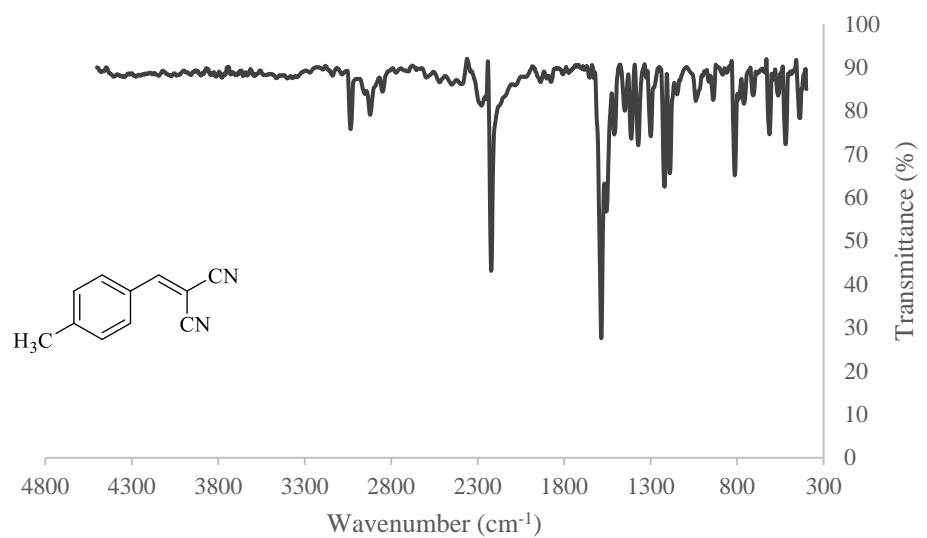
<sup>a</sup> Reaction condition: Benzyl alcohol (1.0 mmol), malononitrile (1.2 mmol), solvent (6.0 mL), Time A (10.0 h), Time B (4.0 h). <sup>b</sup> Isolated yield of B



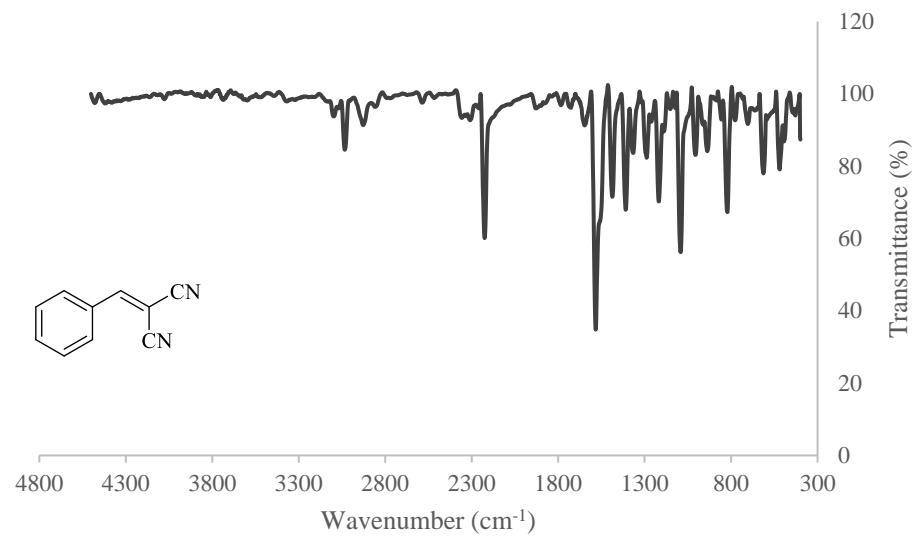
**Figure S6:** FT-IR spectrum of 2-(4-methoxyphenylmethylene)malononitrile



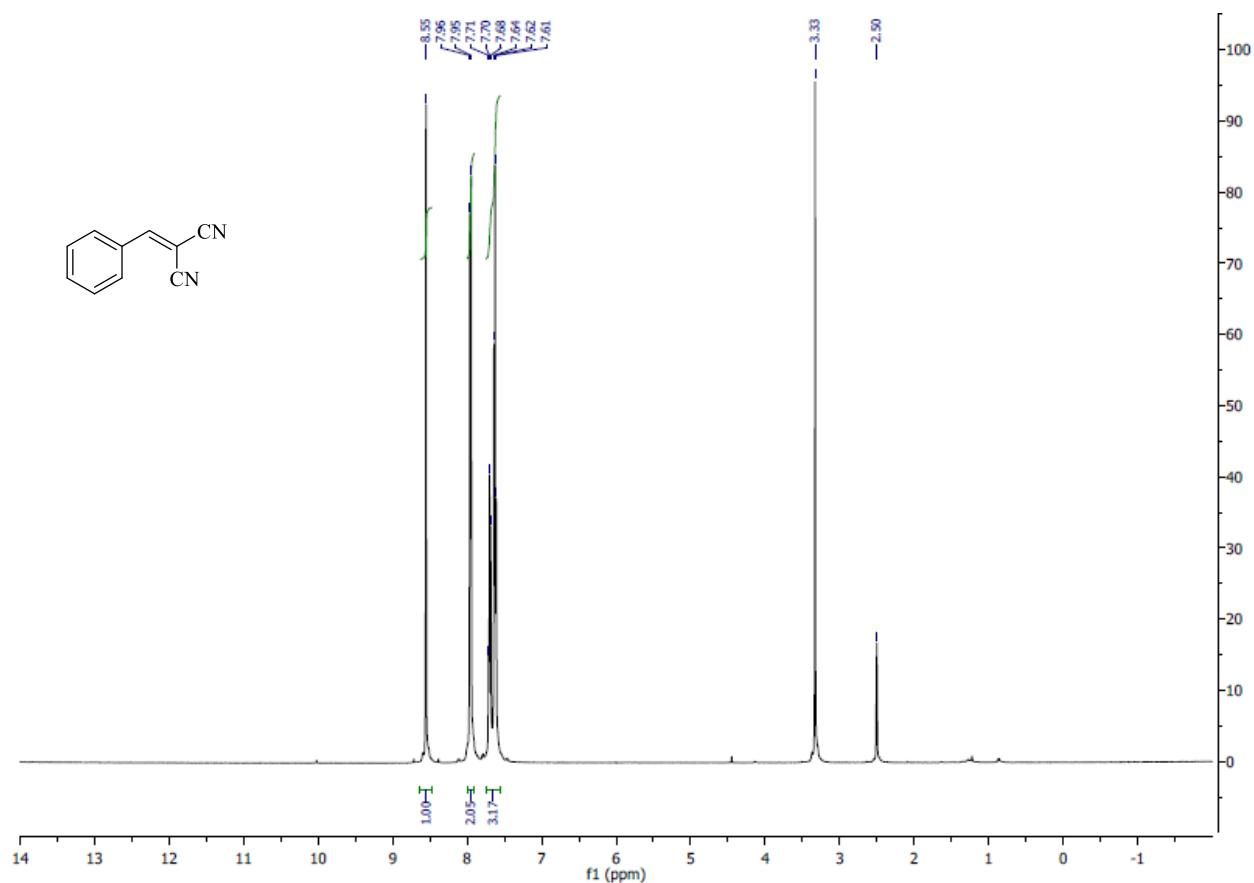
**Figure S7:** FT-IR spectrum of 4-chlorobenzylidenemalononitrile



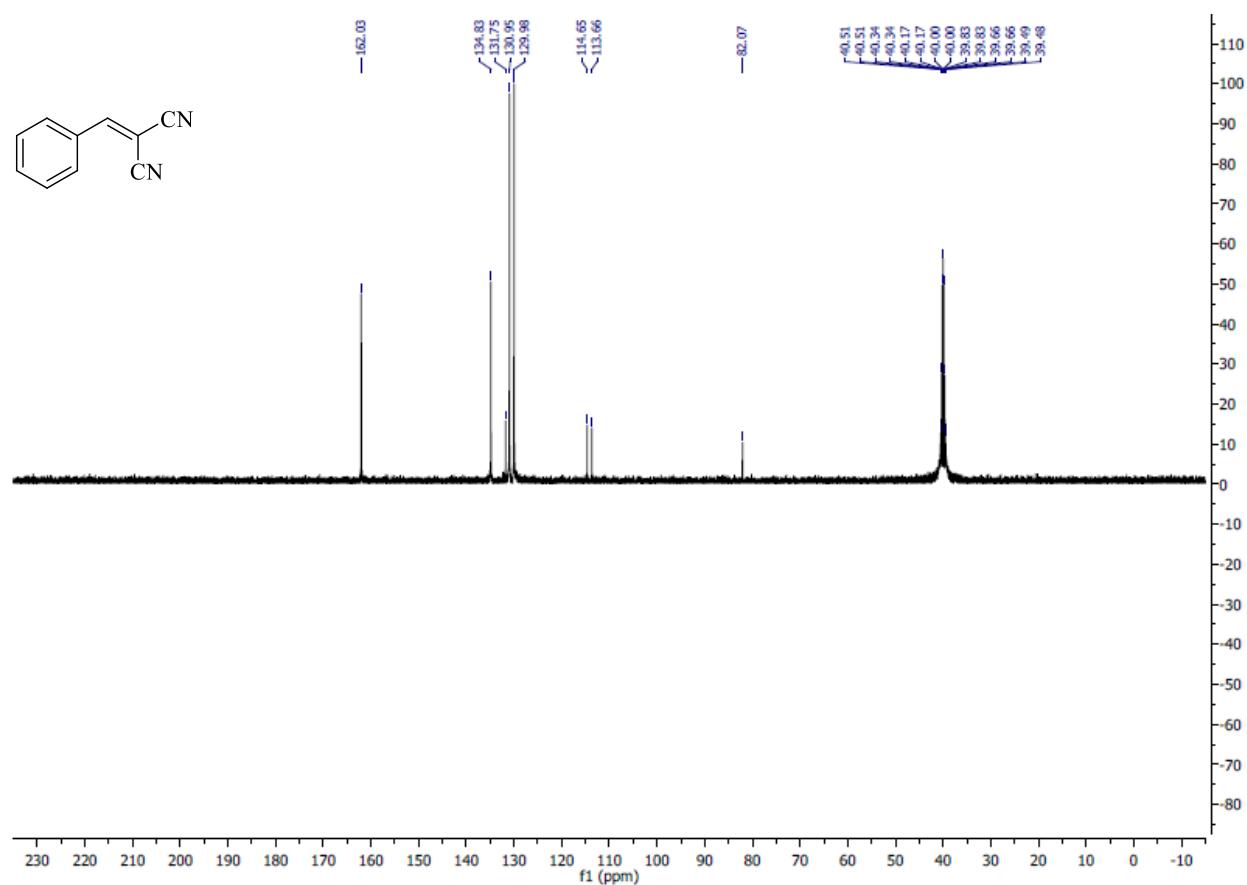
**Figure S8:** FT-IR spectrum of 2-(4-methylbenzylidene)malononitrile



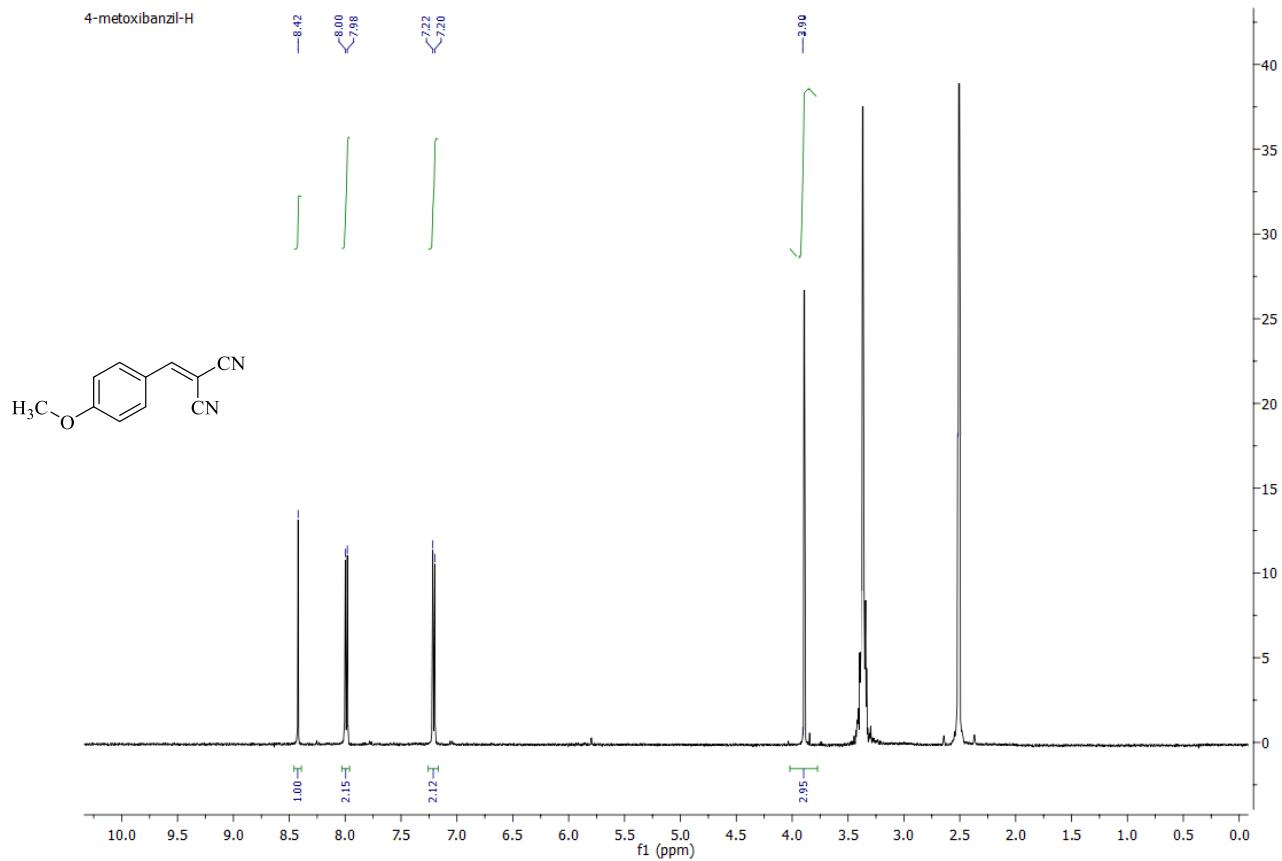
**Figure S9:** FT-IR spectrum of benzylidenemalononitrile



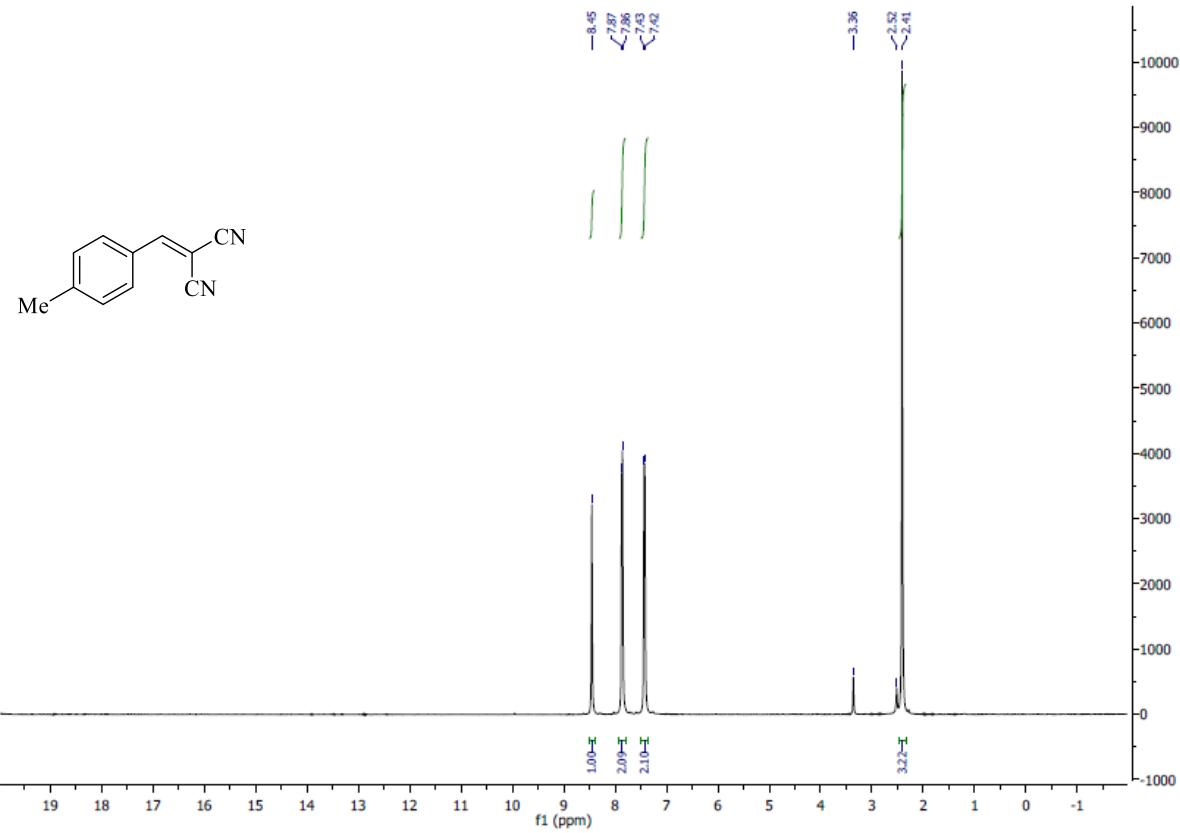
**Figure S10:** <sup>1</sup>H-NMR spectrum of 2-benzylidenemalononitrile



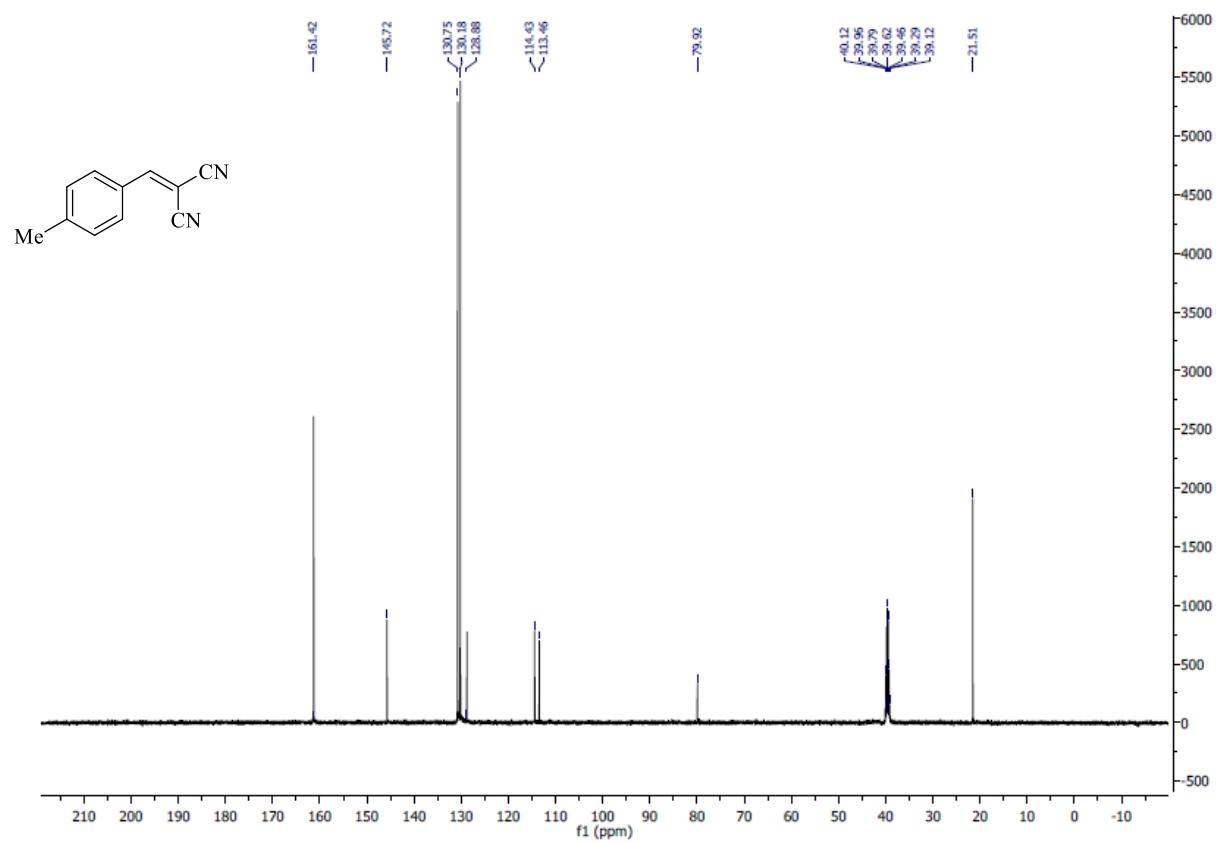
**Figure S11:**  $^{13}\text{C}$ -NMR spectrum of 2-benzylidenemalononitrile



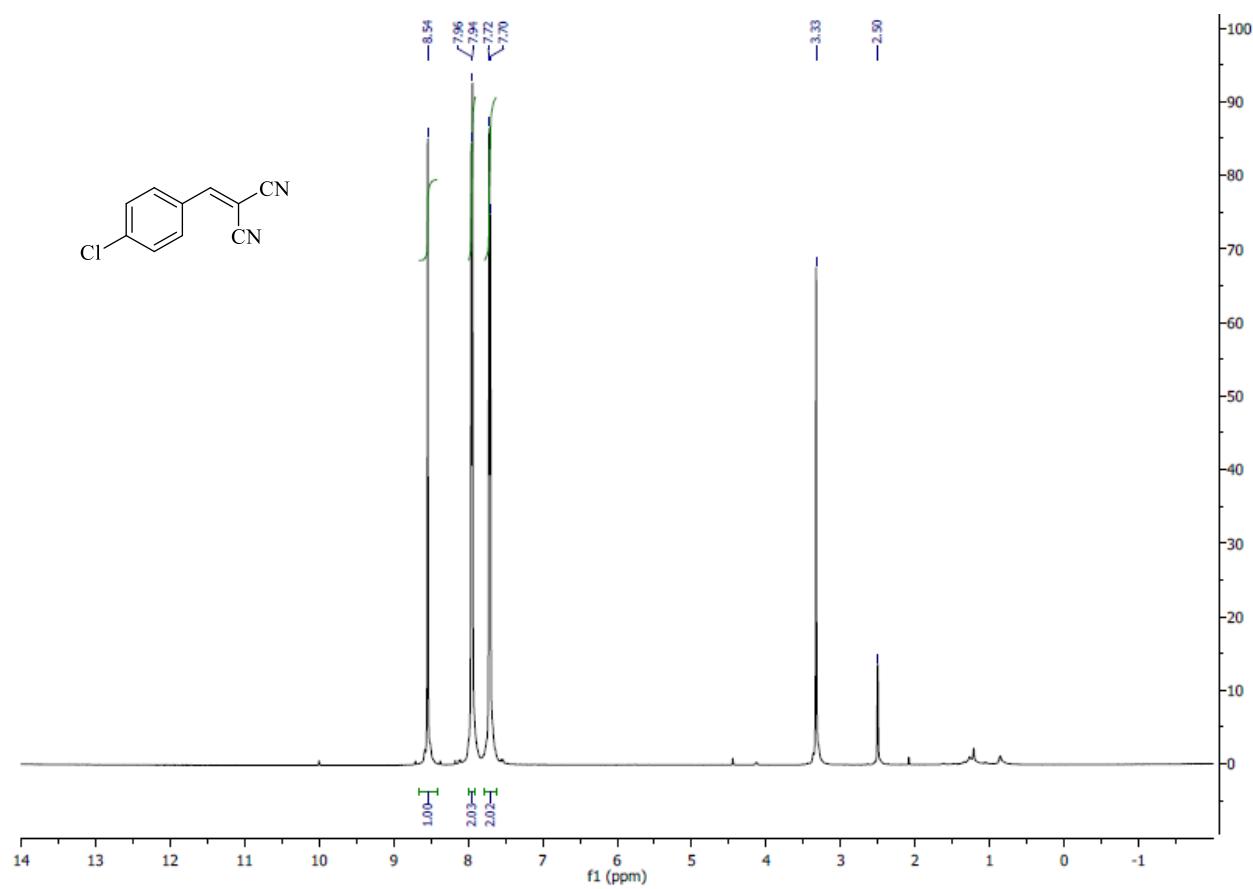
**Figure S12:**  $^1\text{H}$ -NMR spectrum of 2-(4-methoxybenzylidene)malononitrile



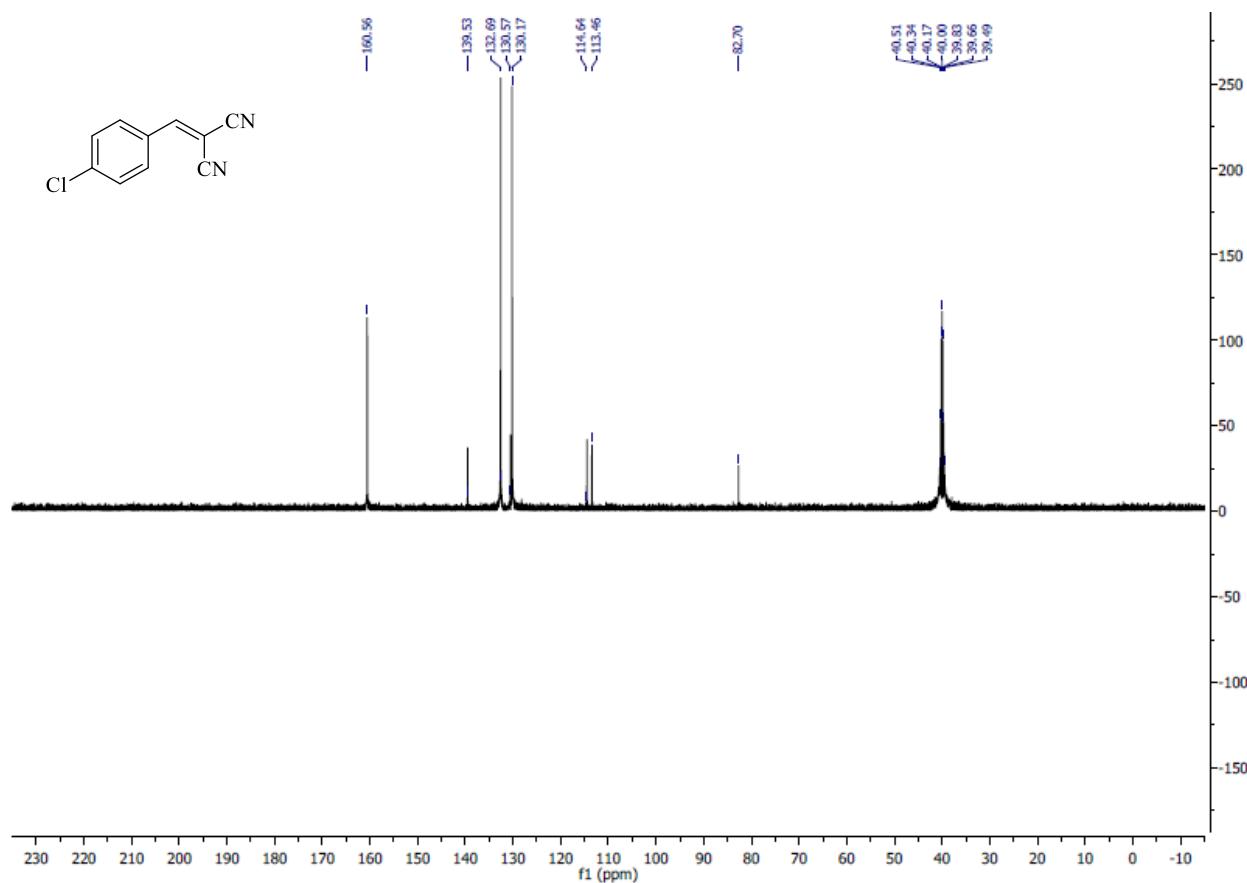
**Figure S13:** <sup>1</sup>H-NMR spectrum of 2-(4-methylbenzylidene)malononitrile



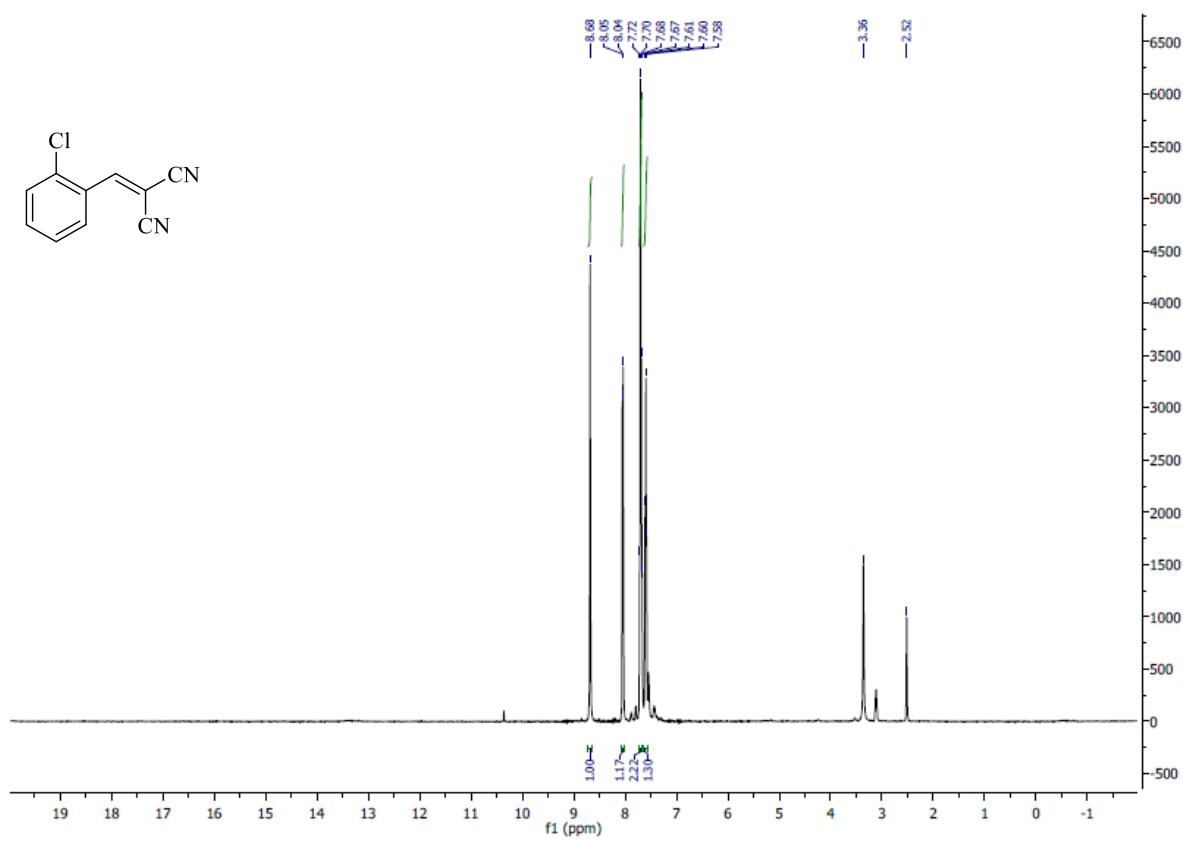
**Figure S14:**  $^{13}\text{C}$ -NMR spectrum of 2-(4-methylbenzylidene)malononitrile



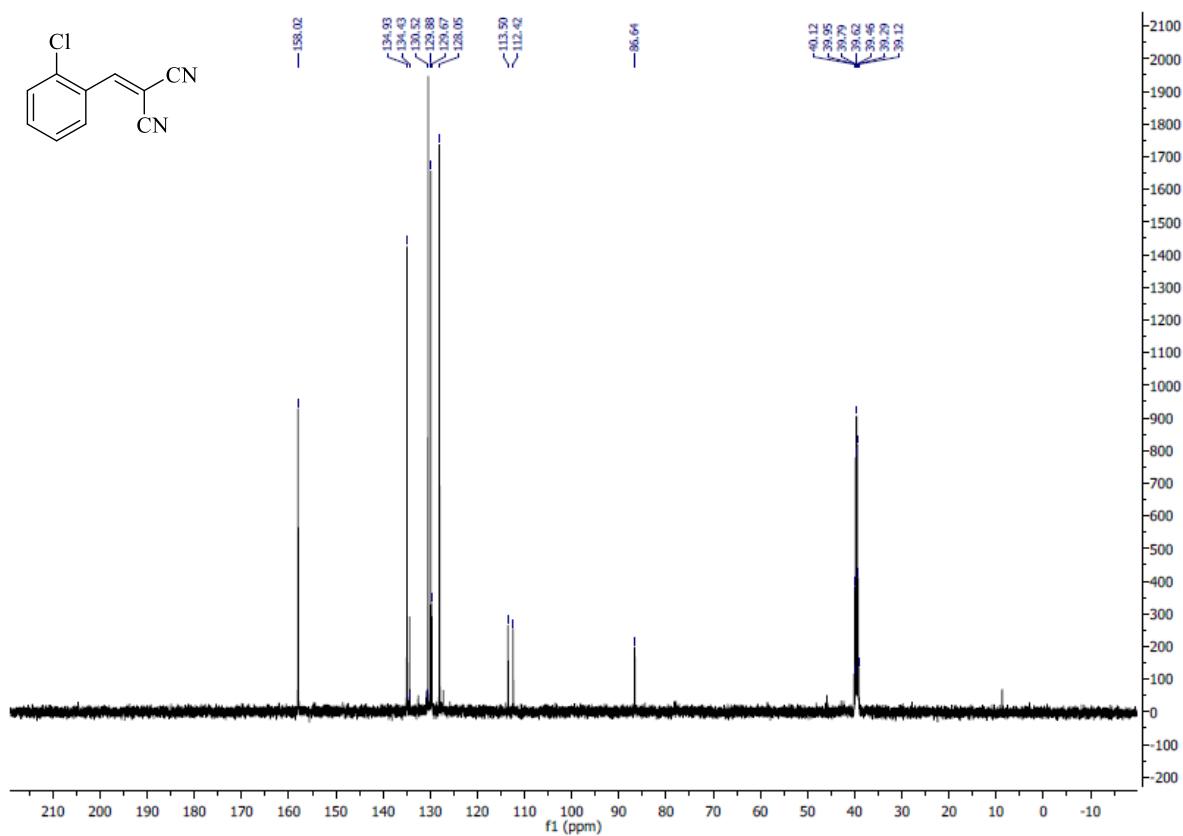
**Figure S15:**  $^1\text{H}$ -NMR spectrum of 2-(4-chlorobenzylidene)malononitrile



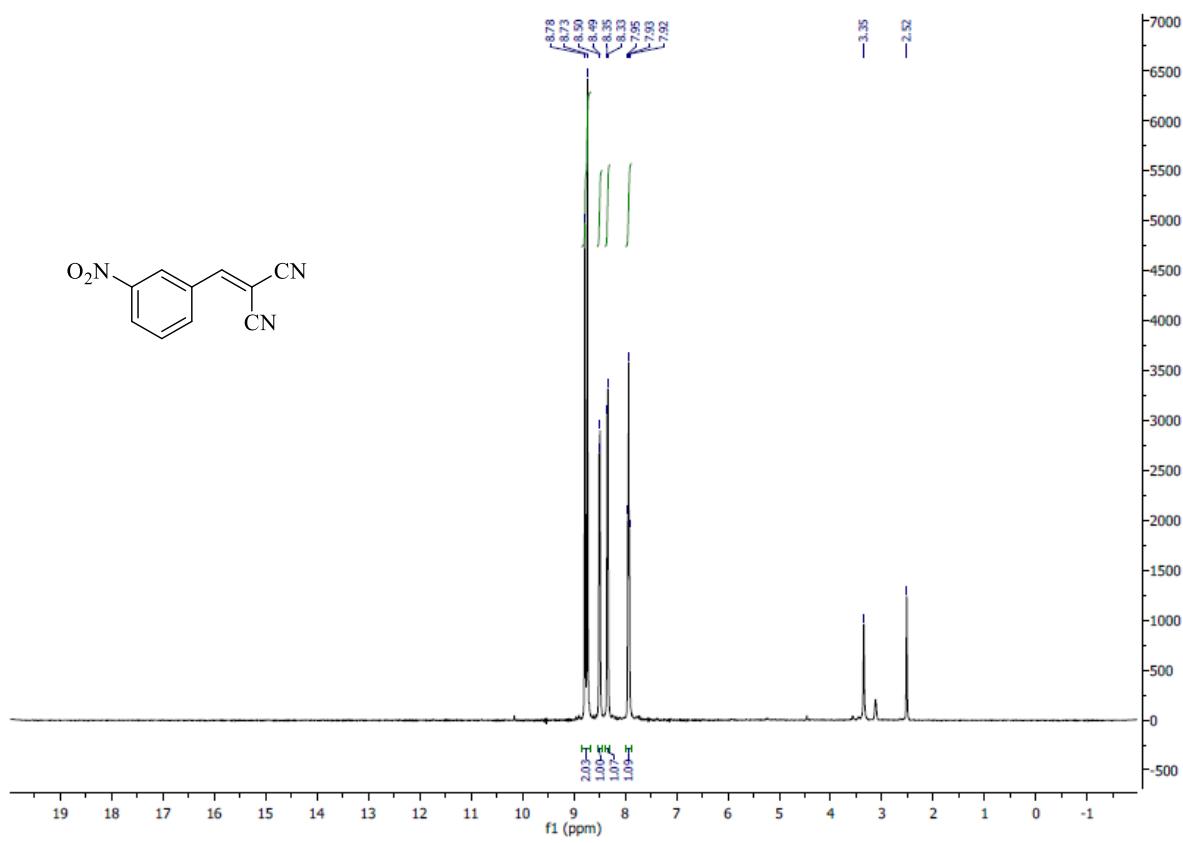
**Figure S16:**  $^{13}\text{C}$ -NMR spectrum of 2-(4-chlorobenzylidene)malononitrile



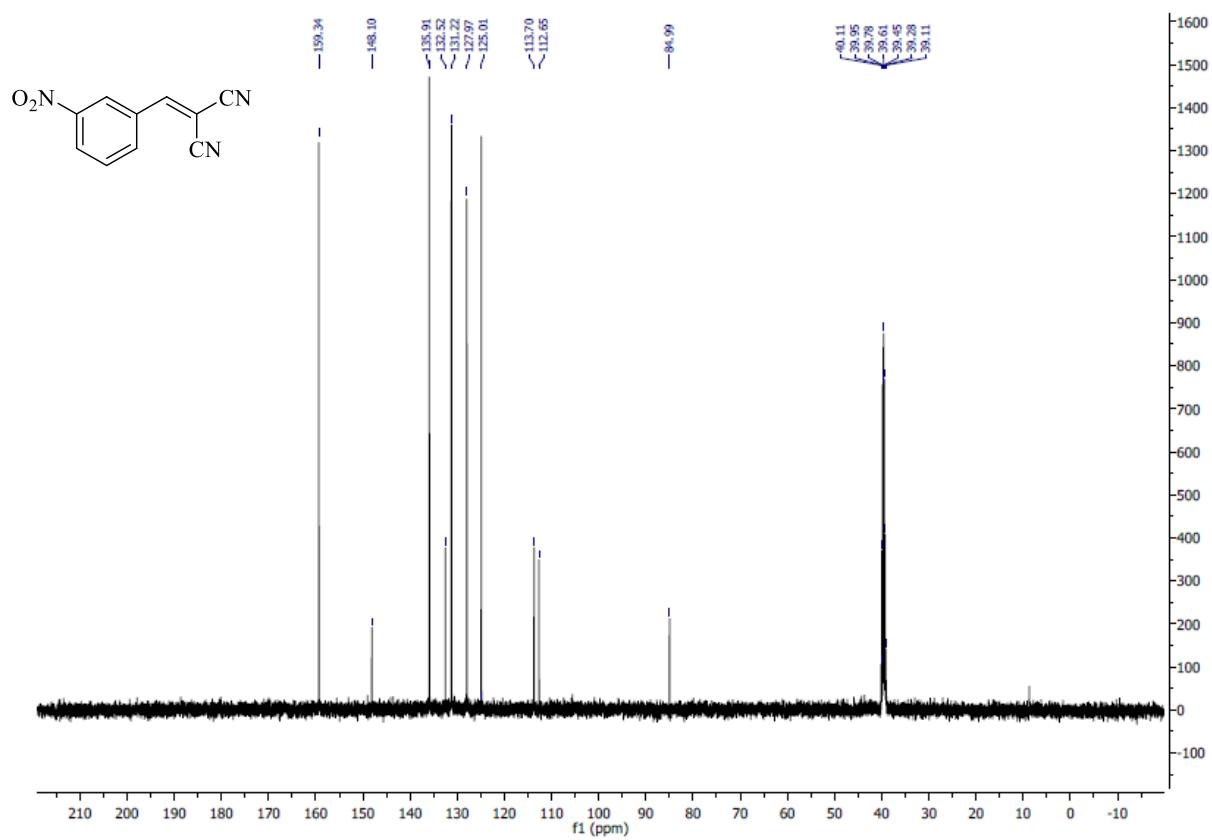
**Figure S17:** <sup>1</sup>H-NMR spectrum of 2-(2-chlorobenzylidene)malononitrile



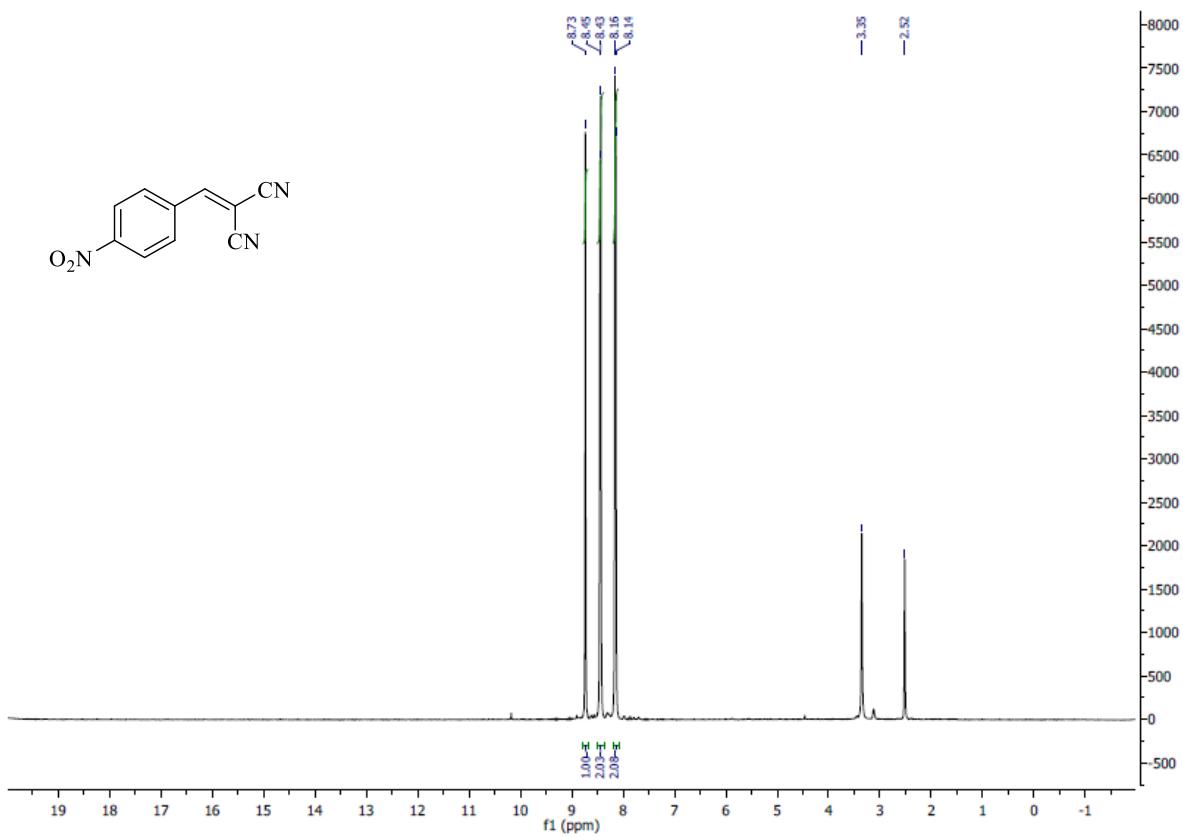
**Figure S18:**  $^{13}\text{C}$ -NMR spectrum of 2-(2-chlorobenzylidene)malononitrile



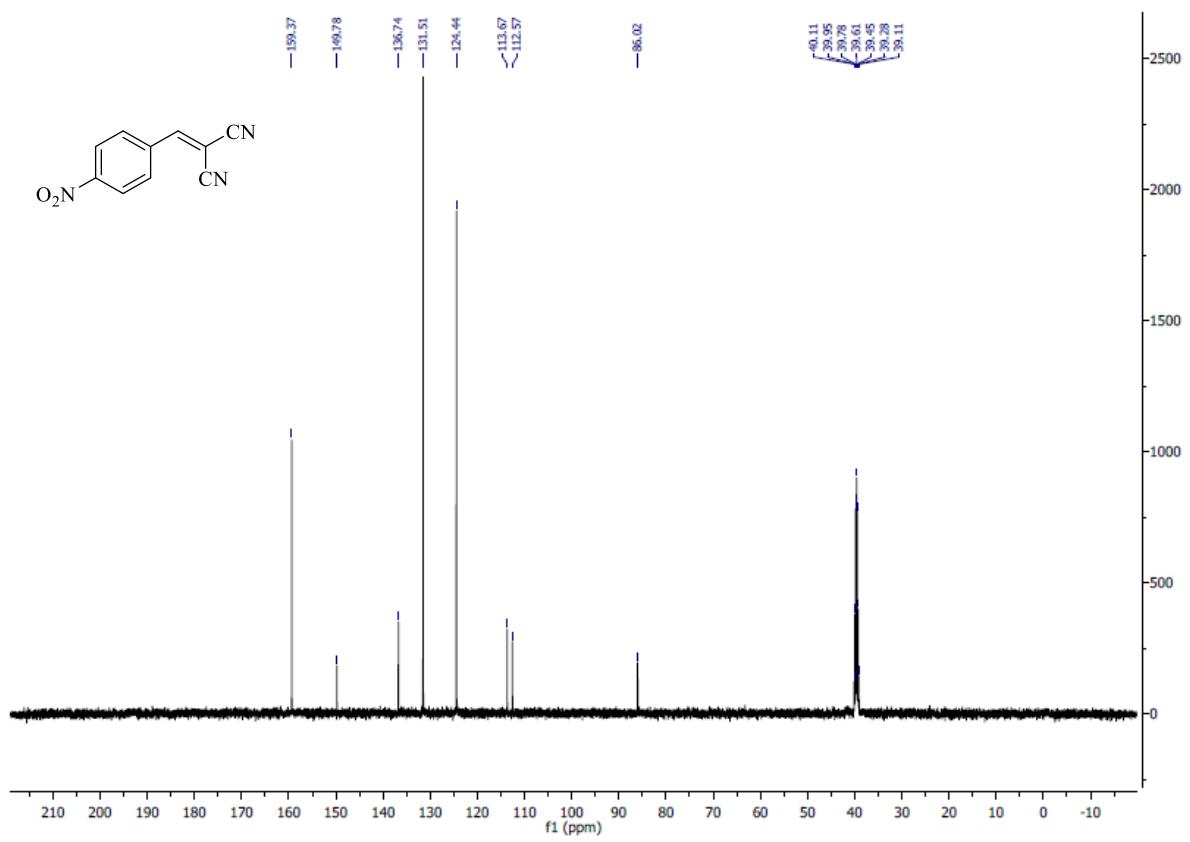
**Figure S19:** <sup>1</sup>H-NMR spectrum of 2-(3-nitrobenzylidene)malononitrile



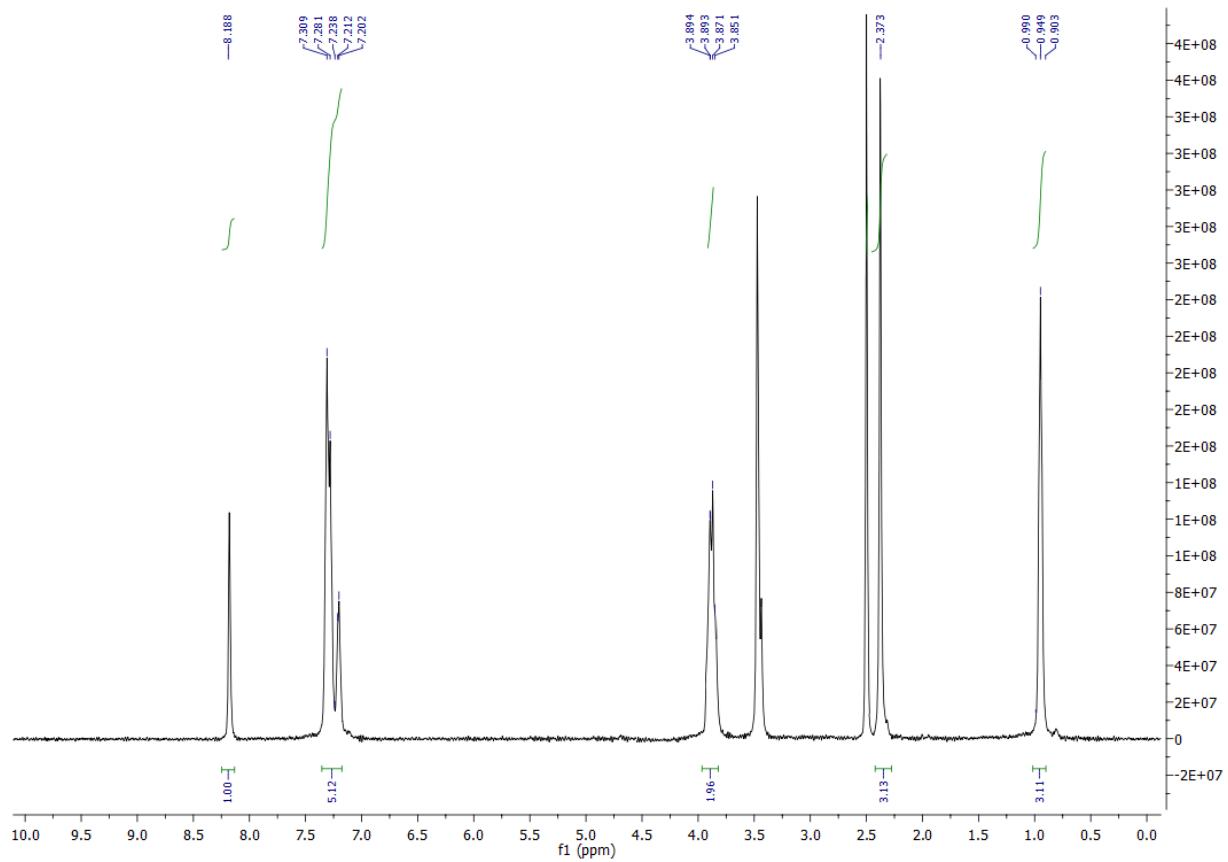
**Figure S20:**  $^{13}\text{C}$ -NMR spectrum of 2-(3-nitrobenzylidene)malononitrile



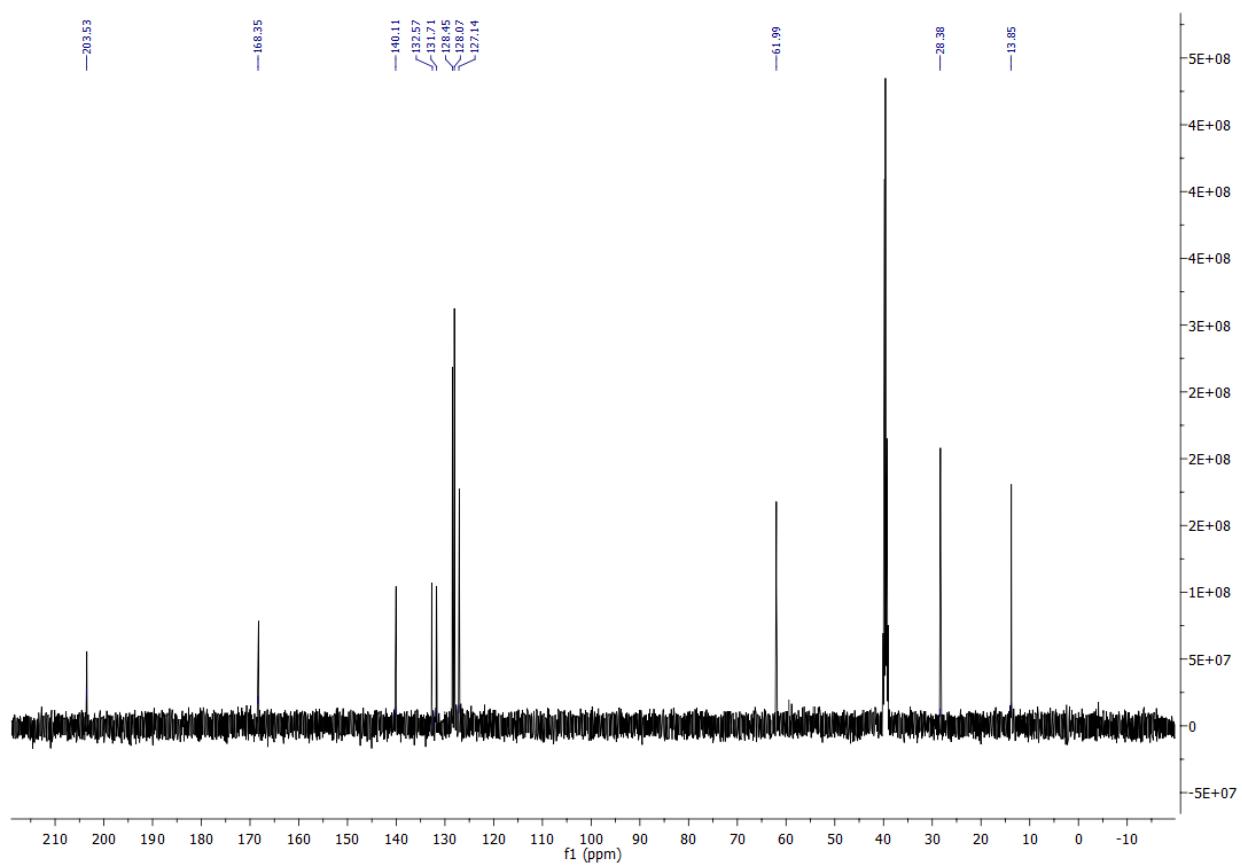
**Figure S21:**  $^1\text{H}$ -NMR spectrum of 2-(4-nitrobenzylidene)malononitrile



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



**Figure S23:**  $^1\text{H}$ -NMR spectrum of ethyl 2-benzylidene-3-oxobutanoate



**Figure S24:**  $^{13}\text{C}$ -NMR spectrum of ethyl 2-benzylidene-3-oxobutanoate