Supporting information

A Brønsted-based deep eutectic solvent as a green catalyst for the sustainable one-pot synthesis of 1,2,4,5-tetrasubstituted imidazole derivatives: *in vitro* cytotoxicity and *in silico* binding studies on HepG2 cells

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Section S1. Chemical and Techniques for analysis

Section S1.1. Chemical

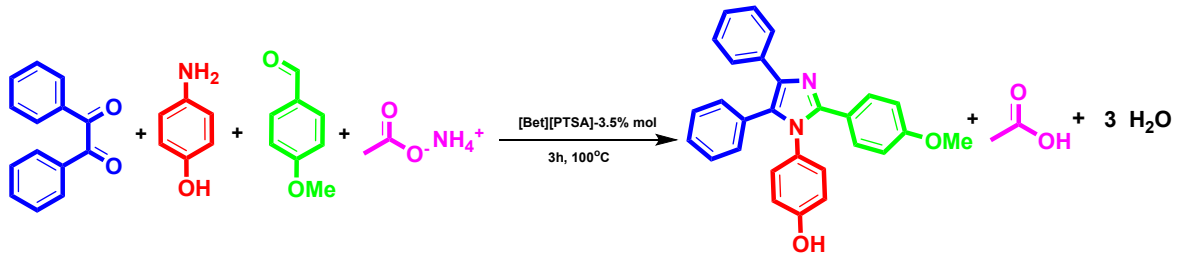
Benzaldehyde (99.5%), 4-methylbenzaldehyde (97%), 4-methoxybenzaldehyde (98%), 4-nitrobenzaldehyde (98%), 4-(dimethylamino)benzaldehyde (for analysis Reag. Ph Eur), betain hydrochloride and TLC silica gel 60 F254 were obtained from Merck. 2-hydroxybenzaldehyde (for synthesis), cyclohexanecarbaldehyde (97%), and vanillin (99%) were obtained from Sigma-Aldrich. 4-bromobenzaldehyde (99%), 4-fluorobenzaldehyde (99%), 3-hydroxybenzaldehyde (98%) and 4-aminophenol (97%) were obtained from Acros. *p*-toluenesulfonic acid (99%) were obtained from Adamas-beta. *n*-hexane (99.5%), methanol (99.5%), ethanol (99.5%), 1,4-dioxane (99.5%), dimethyl sulfoxide (99.5%), acetone (99.5 %), ethyl acetate (99.5%), ammonium acetate (AR), and aniline (99%) were acquired from China.

Section 1.2. Techniques for analysis

Fourier-transform infrared (FT-IR) spectra were obtained using a Jasco FT/IR-6600 type A spectrophotometer. Raman measurements were carried out on a spectrometer equipped with a 532 nm excitation laser and a 1200 lines/mm grating (750 nm blaze), employing a 100× VIS objective, a 100 μm slit width, and a 300 μm confocal aperture. **Thermogravimetric analysis (TGA)** was performed on a TGA Q500 V20.13 Build 39 system under a nitrogen atmosphere, heating samples at a rate of 5 °C per minute. **Nuclear magnetic resonance (NMR) spectroscopy**, including both ^{1}H and ^{13}C spectra, was recorded on a Bruker Advance Neo 500 MHz spectrometer (Bruker BioSpin GmbH) using deuterated solvents. The melting points of the synthesized compounds were measured using a Sanyo-Gallenkamp melting point apparatus.

Section S2. Assessment of green metrics

The green chemistry matrix has been computed for the synthesis of 1-(4-Hydroxyphenyl)-2-(4-methoxyphenyl)-4,5-diphenyl-1*H*-imidazole (**16n**) using the specified parameters:



Scheme S1. Model reaction for green matrix calculation

Table S1. Information about the compounds involved in the reaction.

Compound	M.W. (g/mol)	In the present work M.W. (mg)
Benzil	210.2320	210.2320
4-Aminophenol	109.1280	109,1280
4-Methoxybenzaldehye	136.1500	136.1500
Ammonium acetate	77.0830	77.0830
Compound **16n**	418.4980	418.4980
Acetic acid	60.0520	60.0520
H_2O	18.0150	54.0450

The total mass of reactants = 532.59 mg

Obtained product = 359.91 mg

Environmental factor (E-factor)

$$\text{E-Factor} = \frac{Mass\ of\ waste}{Mass\ of\ product}$$

In which, mass of waste is the mass of water.

$$\text{E-Factor} = \frac{54.0450 + 60.0520}{418.4980} = 0.27$$

(Ideal value of E-factor is considered zero)

Atom-economy (AE)

The optimal value of the AE factor is 100%, indicating that all initial material is fully transformed into the final output.

$$AE = \frac{MW\ of\ product}{\sum MW\ of\ stoichiometric\ reactants} 100$$

$$AE = \frac{418.4980}{210.2320 + 109.1280 + 136.1500 + 77.0830} 100 = 78.58\%$$

Atom efficiency (AEf)

$$AEf = \frac{Yield\ x\ AE}{100\%} 100$$

$$AEf = \frac{86\%\ x\ 78.58\%}{100\%} 100 = 67.57\ \%$$

Process mass intensity (PMI)

$$PMI = \frac{\sum (Mass\ of\ stoichiometric\ reactants\ +\ solvent)}{Mass\ of\ product}$$

$$PMI = \frac{532.59}{359.91} = 1.48$$

Ideal value of PMI = E-Factor + 1 = 0.27 + 1 = 1.27

The variation in value between the findings is rather small.

Reaction mass efficiency (RME)

$$RME = \frac{Mass\ of\ product}{\sum (mass\ of\ stoichiometric\ reactants)} 100\%$$

$$RME = \frac{359.91}{532.59} 100 = 67.58\%$$

Carbon efficiency (CE) (%)

$$CE = \frac{Amount\ of\ carbon\ in\ product}{Total\ carbon\ present\ in\ reactant} 100\%$$

$$CE = \frac{0.86\ x\ 28}{14 + 6 + 8 + 2} 100 = 80.3\%$$

Eco-score (E-score)

Ideal reactions: Eco-score value is 100.

Eco-scale from 0 to 100 using the following scores: > 75, excellent; > 50, acceptable; and < 50, inadequate. E-score has been calculated for the reaction based on the following 6 parameters below.

Table S2. Parameters for calculating the eco-score.

Entry	Parameter	Values	Penalty points
1	Yield	(100-86)/2	7
2	Price of the reaction component	Inexpensive	0.0
3	Safety (Reactant)	T= 5 + 5 + 5 + 5 = 20	20.0
4	Technical setup	Common setup	0.0
5	Temperature /time	100/ 3 h	3.0
6	Workup and purification	Crystallization	1.0
	Total penalty points		31.0
	Based on the hazard warning symbols		

Eco-Score = 100 – (The sum of individual penalties) = 100 – 31 = 69 (> 50, acceptable synthesis).

As per the above results, it was concluded that the reaction has an extremely low Environment-factor (E-factor = 0.27), high atom economy (AE = 78.58%), medium atom efficiency (AEf = 67.57%), low process mass intensity (PMI = 1.48), good carbon efficiency (CE = 80.3%), and medium reaction mass efficiency (RME = 67.58%), with acceptable eco-score (69%). These values clearly indicate the eco-friendliness of the present synthesis.

Section S3. Binding energy and interaction

Table S3: Binding energy and interaction with the five poses of 15 compounds **16a-o**

Compound	Binding energy (kcal/mol)[a]	Interacting residues
16a	**-7.66**	
	-7.55	
	-7.51	Asn288
	-7.35	Trp388
	-7.11	
16b	**-7.63**	
	-7.58	
	-7.44	Trp412
	-7.30	
	-7.25	Asn411
16c	**-7.57**	Asn411
	-7.48	
	-7.36	Asn412
	-7.29	
	-7.23	Asn317
16d	**-8.23**	Asn288
	-8.01	Trp388
	-7.73	
	-7.71	
	-7.67	
16e	**-8.07**	
	-7.96	
	-7.95	Trp412
	-7.80	
	-7.78	
16f	**-8.16**	
	-8.08	
	-8.03	
	-7.90	

Compound	Binding energy (kcal/mol)[a]	Interacting residues
	-7.79	Trp388
16g	**-7.89**	
	-7.85	
	-7.42	
	-7.42	
	-7.40	
16h	**-7.90**	
	-7.79	
	-7.73	
	-7.59	
	-7.54	
16i	**-7.84**	
	-7.45	Glu380
	-7.27	Glu380
	-7.10	Glu380
	-7.10	
16j	**-8.33**	
	-8.17	
	-8.12	
	-8.12	
	-8.01	
16k	**-7.81**	
	-7.78	
	-7.59	Glu380
	-7.46	Glu380
	-7.40	
16l	**-7.74**	
	-7.68	Glu380
	-7.67	
	-7.62	Trp412
	-7.48	Glu380

Compound	Binding energy (kcal/mol)[a]	Interacting residues
16m	**-7.64**	
	-7.49	Glu380
	-7.42	
	-7.40	
	-7.33	Glu380
16n	**-8.23**	Asn288
	-8.22	
	-8.18	
	-8.16	
	-7.80	
16o	**-8.01**	
	-7.92	Thr137
	-7.89	
	-7.75	Trp412
	-7.71	Trp388

[a]The binding energy value selected for the molecular dynamics simulation is highlighted in bold.

```
Found 7 hbonds.
donor            acceptor         occupancy
THR137-Side      *0-Main          0.01%
TRP388-Side      *0-Main          0.40%
GLN282-Side      *0-Main          0.07%
*0-Main          THR137-Side      0.23%
TRP412-Side      *0-Main          1.66%
GLN283-Side      *0-Main          0.02%
GLN161-Side      *0-Main          0.04%
```

Fig. S1. Analysis of hydrogen in molecular dynamic **16d.**

Found 9 hbonds.

donor	acceptor	occupancy
*0-Main-O	THR30-Side-OG1	47.96%
GLN282-Side-NE2	*0-Main-N	0.03%
TYR292-Side-OH	*0-Main-O	2.94%
TRP388-Side-NE1	*0-Main-N	3.01%
THR137-Side-OG1	*0-Main-O	0.01%
TRP412-Side-NE1	*0-Main-O	0.17%
GLN161-Side-NE2	*0-Main-C	0.02%
*0-Main-O	ASN415-Side-OD1	16.80%
SER80-Side-OG	*0-Main-O	0.01%

Fig. S2. Analysis of hydrogen in molecular dynamic **16j.**

Found 10 hbonds.

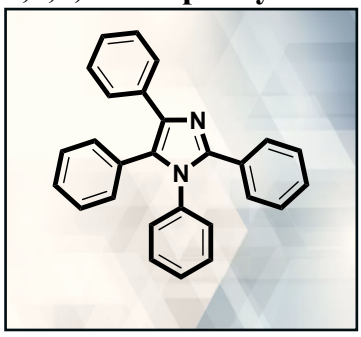
donor	acceptor	occupancy
TRP388-Side-NE1	*0-Main-N	1.78%
*0-Main-O	THR30-Side-OG1	58.80%
THR137-Side-OG1	*0-Main-O	0.91%
GLN282-Side-NE2	*0-Main-N	0.01%
TRP412-Side-NE1	*0-Main-O	0.20%
*0-Main-O	TYR292-Side-OH	1.09%
GLN161-Side-NE2	*0-Main-C	0.30%
*0-Main-O	ASN415-Side-OD1	0.20%
THR137-Side-OG1	*0-Main-C	0.05%
TYR292-Side-OH	*0-Main-O	0.05%

Fig. S3. Analysis of hydrogen in molecular dynamic **16n.**

Section S4. Spectra data

The synthetic procedure is described in the manuscript. The derivatives were prepared from benzaldehyde derivatives, aniline derivatives, benzil, and ammonium acetate, and are designated as **16a-16o**.

1,2,4,5-tetraphenyl-1*H*-imidazole (16a)



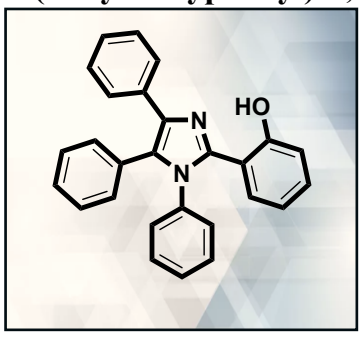
The actual weighed amounts used in the synthesis of compound **16a** are as follows: benzaldehyde (0.1069 g), aniline (0.0941 g), benzil (0.2107 g) and ammonium acetate (0.0788 g).

The white solid was obtained with 75% (279 mg) and M.p. = 215-217 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 7.51 (d, J = 7.5 Hz, 2H), 7.40-7.38 (m, 2H), 7.33-7.32 (m, 3H), 7.30-7.28 (m, 6H), 7.26-7.23 (m, 6H), 7.19-7.16 (m, 1H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 146.0, 136.8, 136.6, 134.4, 131.3, 131.1, 130.4, 129.1, 128.7, 128.7, 128.4, 128.4, 128.3, 128.2, 128.2, 128.1, 126.4, 126.3 ppm.

2-(2-Hydroxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (16b)



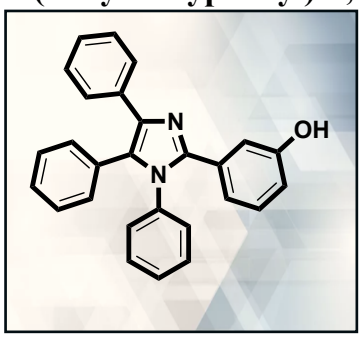
The actual weighed amounts used in the synthesis of compound **16b** are as follows: 2-hydroxybenzaldehyde (0.122 g), aniline (0.0949 g), benzil (0.2111 g) and ammonium acetate (0.7781 g).

The white solid was obtained with 51% (198 mg) and M.p. = 255-259 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 12.60 (s, 1H), 7.45 (d, J = 7.5 Hz, 2H), 7.39-7.36 (m, 5H), 7.31-7.27 (m, 7H), 7.23-7.20 (m, 1H), 7.19-7.15 (m, 1H), 6.95 (d, J = 8.5 Hz, 1H), 6.67 (dd, J = 8.0, 1.0 Hz, 1H), 6.54 (t, J = 7.5 Hz, 1H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.3, 144.4, 136.5, 134.4, 133.1, 131.2, 130.7, 130.1, 129.6, 129.3, 129.2, 128.7, 128.6, 128.5, 128.4, 126.9, 126.7, 126.1, 118.1, 116.9, 113.8 ppm.

2-(3-Hydroxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (16c)



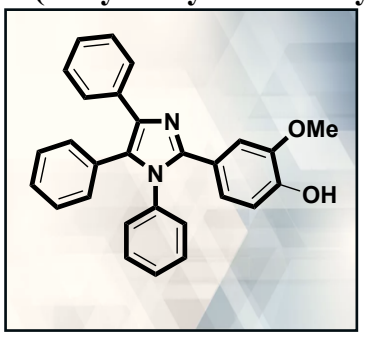
The actual weighed amounts used in the synthesis of compound 16c are as follows: 3-hydroxylbenzaldehyde (0.1231 g), aniline (0.0949 g), benzil (0.2100 g) and ammonium acetate (0.0790 g).

The white solid was obtained with 57% (221 mg) and M.p. = 205-209 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.50 (s, 1H), 7.48 (d, J = 7.0 Hz, 2H), 7.32-7.28 (m, 6H), 7.25-7.22 (m, 6H), 7.18-7.15 (m, 1H), 7.02 (t, J = 8.0 Hz, 1H), 6.95 (s, 1H), 6.70-6.66 (m, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.0, 146.0, 136.7, 136.7, 134.4, 131.5, 131.2, 131.1, 130.4, 129.1, 129.0, 128.7, 128.4, 128.4, 128.1, 126.4, 126.3, 118.9, 115.4, 115.4 ppm.

2-(4-Hydroxy-3-methoxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (16d)



The actual weighed amounts used in the synthesis of compound **16d** are as follows: vaniline (0.1523 g), aniline (0.0941 g), benzil (0.2110 g) and ammonium acetate (0.0779 g).

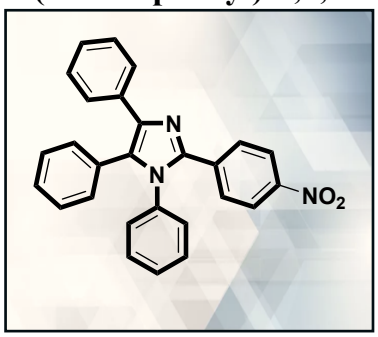
The white solid was obtained with 45% (188 mg) and M.p. = 211-216 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.25 (s, 1H), 7.48 (d, J = 7.5 Hz, 2H), 7.35-7.33 (m, 3H), 7.28 (dd, J = 6.0, 3.5 Hz, 3H), 7.25-7.22 (m, 6H), 7.18-7.15 (m, 1H), 6.86 (dd, J = 8.5, 2.0 Hz, 1H), 6.83 (d, J = 2.0 Hz, 1H), 6.67 (d, J = 8.0 Hz, 1H), 3.50 (s, 3H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 146.9, 146.8, 146.3, 137.0, 136.4, 134.5, 131.1, 130.6,

130.6, 129.1, 128.9, 128.5, 128.4, 128.2, 128.1, 126.4, 126.3, 121.4, 115.1, 112.4, 55.1 ppm.

2-(4-Nitrophenyl)-1,4,5-triphenyl-1*H*-imidazole (16e)



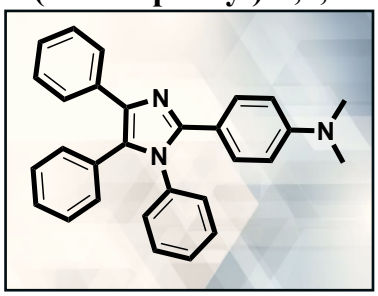
The actual weighed amounts used in the synthesis of compound **16e** are as follows: 4-nitrobenzaldehyde (0.1510 g), aniline (0.0948 g), benzil (0.2100 g) and ammonium acetate (0.0790 g).

The pale orange solid was obtained with 60% (250 mg) and M.p. = 219-220 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 8.14 (d, J = 9.0 Hz, 2H), 7.62 (d, J = 8.5 Hz, 2H), 7.52 (d, J = 7.5 Hz, 2H), 7.38 (d, J = 6.5 Hz, 3H), 7.34-7.31 (m, 5H), 7.28-7.25 (m, 4H), 7.20 (t, J = 7.0 Hz, 1H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 146.6, 143.8, 137.8, 136.3, 136.2, 133.9, 132.8, 131.1, 129.8, 129.4, 129.2, 128.8, 128.7, 128.6, 128.5, 128.2, 126.8, 126.4, 123.5 ppm.

2-(4-Nitrophenyl)-1,4,5-triphenyl-1*H*-imidazole (16f)



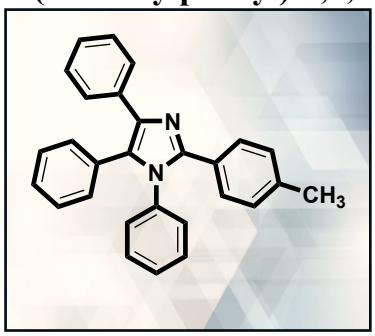
The actual weighed amounts used in the synthesis of compound **16f** are as follows: 4-dimethylamino benzaldehyde (0.1492 g), aniline (0.0931 g), benzil (0.2109 g) and ammonium acetate (0.0811 g).

The gray solid was obtained with 61% (253 mg) and M.p. = 209-212 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 7.47 (d, J = 7.0 Hz, 2H), 7.34-7.32 (m, 3H), 7.28-7.26 (m, 3H), 7.24-7.14 (m, 9H), 6.57 (d, J = 10.0 Hz, 2H), 2.87 (s, 6H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 149.9, 146.7, 137.1, 136.3, 134.7, 131.1, 130.7, 130.4, 129.1, 129.0, 128.8, 128.5, 128.3, 128.1, 128.0, 126.3, 126.2, 117.7, 111.3 ppm.

2-(4-Methylphenyl)-1,4,5-triphenyl-1*H*-imidazole (16g)



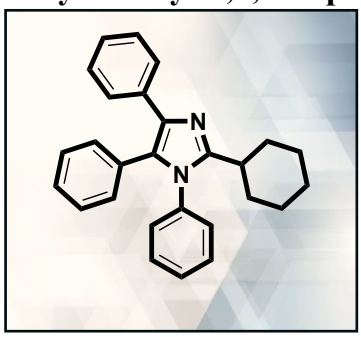
The actual weighed amounts used in the synthesis of compound **16g** are as follows: 4-methylbenzaldehyde (0.121 g), aniline (0.0945 g), benzil (0.2100 g) and ammonium acetate (0.0773 g).

The white solid was obtained with 65% (251 mg) and M.p. = 189-192 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 7.49 (d, J = 8.0 Hz, 2H), 7.32-7.31 (m, 3H), 7.29-7.27 (m, 4H), 7.26 (s, 2H), 7.24-7.22 (m, 5H), 7.17 (t, J = 7.5 Hz, 1H), 7.08 (d, J = 8.0 Hz, 2H), 2.26 (s, 3H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 146.1, 137.8, 136.7, 136.7, 134.4, 131.1, 131.0, 130.4, 129.1, 128.7, 128.7, 128.6, 128.4, 128.3, 128.1, 128.1, 127.6, 126.4, 126.3, 20.7 ppm.

2-Cyclohexyl-1,4,5-triphenyl-1*H*-imidazole (16h)



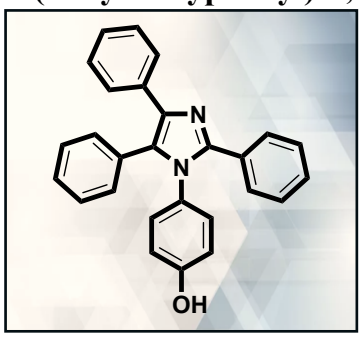
The actual weighed amounts used in the synthesis of compound **16h** are as follows: cyclohexanecarbadehyde (0.1123 g), aniline (0.0934 g), benzil (0.2110 g) and ammonium acetate (0.0781 g).

The white solid was obtained with 46% (174 mg) and M.p. = 206-209 °C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 7.44 (d, J = 7.5 Hz, 2H), 7.40-7.35 (m, 3H), 7.29 (d, J = 7.5 Hz, 2H), 7.24-7.21 (m, 3H), 7.20-7.20 (m, 5H), 2.41 (t, J = 11.5 Hz, 1H), 1.81 (d, J = 12.0 Hz, 2H), 1.73-1.69 (m, 2H), 1.66-1.58 (m, 3H), 1.22 (dd, J = 25.0, 12.5 Hz, 1H), 1.08 (dd, J = 25.0, 12.5 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 152.0, 135.9, 135.4, 134.9, 130.8, 129.1, 128.8, 128.6, 128.4, 128.3, 128.0, 127.9, 126.2, 126.0, 35.5, 31.7, 25.6, 25.4 ppm.

1-(4-Hydroxyphenyl)-2,4,5-triphenyl-1*H*-imidazole (16i)



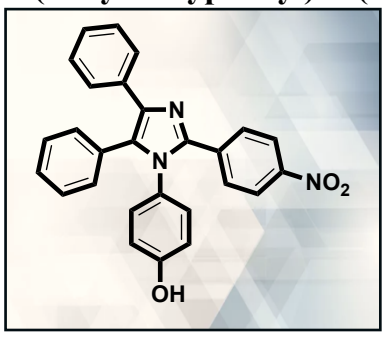
The actual weighed amounts used in the synthesis of compound **16i** are as follows: benzaldehyde (0.1060 g), 4-aminophenol (0.1091 g), benzil (0.2100 g) and ammonium acetate (0.0771 g).

The white to pale gray solid was obtained with 70% (272 mg) and M.p. = 301-306 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.75 (s, 1H), 7.49 (d, J = 7.5 Hz, 2H), 7.44-7.43 (m, 2H), 7.30 (dd, J = 6.5, 4.0 Hz, 6H), 7.25-7.22 (m, 4H), 7.18-7.15 (m, 1H), 7.05 (d, J = 6.0 Hz, 2H), 6.67 (d, J = 8.5 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.3, 146.1, 136.6, 134.5, 131.6, 131.1, 130.6, 130.6, 129.8, 128.4, 128.2, 128.1, 128.1, 127.8, 126.3, 115.6 ppm.

1-(4-Hydroxyphenyl)-2-(4-nitrophenyl)-4,5-diphenyl-1*H*-imidazole (16j)



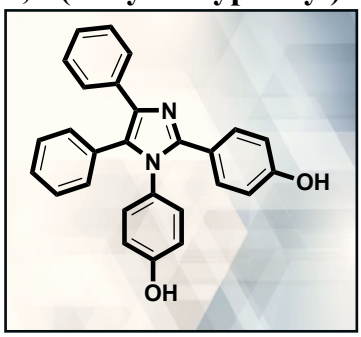
The actual weighed amounts used in the synthesis of compound **16j** are as follows: 4-nitrobenzaldehyde (0.1511 g), 4-aminophenol (0.1092 g), benzil (0.2100 g) and ammonium acetate (0.0781 g).

The brownish-orange solid was obtained with 65% (282 mg) and M.p. = 296-301 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.85 (s, 1H), 8.17 (d, J = 9.0 Hz, 2H), 7.67 (d, J = 9.0 Hz, 2H), 7.50 (d, J = 7.5 Hz, 2H), 7.33 (t, J = 3.0 Hz, 3H), 7.27-7.24 (m, 4H), 7.21-7.18 (m, 1H), 7.12 (d, J = 8.5 Hz, 2H), 6.71 (dd, J = 6.5, 2.0 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.7, 146.5, 143.9, 137.6, 136.5, 134.0, 133.1, 131.1, 130.1, 129.7, 128.6, 128.5, 128.5, 128.4, 128.2, 127.3, 126.7, 126.4, 123.5, 115.9 ppm.

1,2-(4-Hydroxyphenyl)-4,5-diphenyl-1*H*-imidazole (16k)



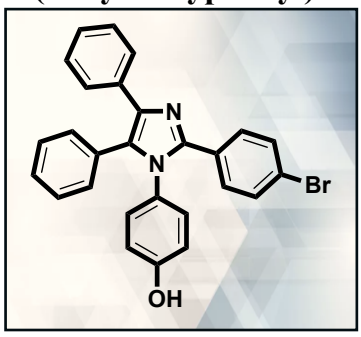
The actual weighed amounts used in the synthesis of compound **16k** are as follows: 4-hydroxylbenzaldehyde (0.1221 g), 4-aminophenol (0.1099 g), benzil (0.2117 g) and ammonium acetate (0.0790 g).

The white solid was obtained with 69% (279 mg) and M.p. = 335-337 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.68 (br, 2H), 7.47 (dd, J = 8.5, 1.0 Hz, 2H), 7.30-7.28 (m, 3H), 7.24-7.20 (m, 6H), 7.16-7.13 (m, 1H), 7.00 (dd, J = 6.5, 2.5 Hz, 2H), 6.66 (td, J = 6.5, 2.5 Hz, 4H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.5, 157.1, 146.5, 136.1, 134.7, 131.1, 130.9, 129.8, 129.6, 128.3, 128.1, 128.0, 126.3, 126.1, 121.5, 115.5, 114.9 ppm.

1-(4-Hydroxyphenyl)-2-(4-bromophenyl)-4,5-diphenyl-1*H*-imidazole (16l)



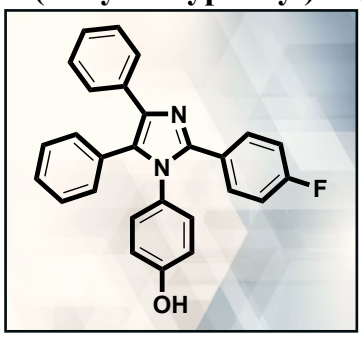
The actual weighed amounts used in the synthesis of compound **16l** are as follows: 4-bromobenzaldehyde (0.1855 g), 4-aminophenol (0.110 g), benzil (0.2101 g) and ammonium acetate (0.0788 g).

The grey pale solid was obtained with 52% (243 mg) and M.p. = 320-323 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.78 (s, 1H), 7.52 (d, J = 8.5 Hz, 2H), 7.48 (d, J = 7.0 Hz, 2H), 7.35 (dd, J = 6.5, 2.0 Hz, 2H), 7.31-7.30 (m, 3H), 7.25-7.20 (m, 4H), 7.18-7.15 (m, 1H), 7.06 (dd, J = 6.5, 2.0 Hz, 2H), 6.67 (dd, J = 6.5, 2.5 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.4, 145.0, 136.8, 134.3, 131.9, 131.1, 131.1, 130.4, 129.9, 129.7, 129.7, 128.4, 128.3, 128.1, 127.5, 126.4, 126.3, 121.7, 115.7 ppm.

1-(4-Hydroxyphenyl)-2-(4-flourophenyl)-4,5-diphenyl-1*H*-imidazole (16m)



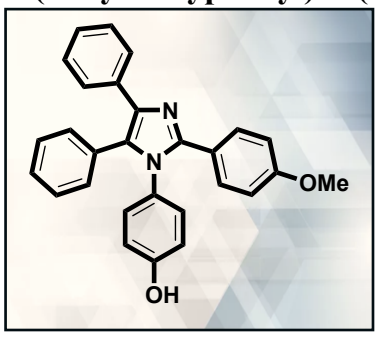
The actual weighed amounts used in the synthesis of compound **16m** are as follows: 4-fluorobenzaldehyde (0.1251 g), 4-aminophenol (0.1087 g), benzil (0.2121 g) and ammonium acetate (0.0791 g).

The white solid was obtained with 10% (41 mg) and M.p. = 298-300 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.76 (s, 1H), 7.48-7.44 (m, 4H), 7.32-7.30 (m, 3H), 7.25-7.22 (m, 4H), 7.16 (t, J = 8.5 Hz, 3H), 7.06 (d, J = 8.5 Hz, 2H), 6.66 (d, J = 8.0 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.3, 145.2, 136.5, 134.4, 131.5, 131.1, 130.5, 130.2, 130.2, 129.8, 128.4, 128.3, 128.4, 128.3, 128.1, 127.6, 126.3, 115.6, 115.2, 115.0 ppm.

1-(4-Hydroxyphenyl)-2-(4-methoxyphenyl)-4,5-diphenyl-1*H*-imidazole (16n)



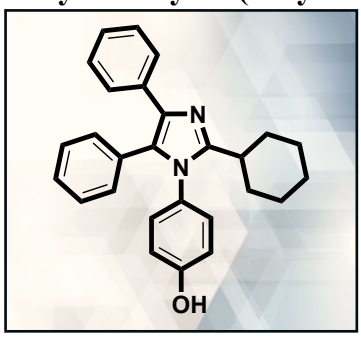
The actual weighed amounts used in the synthesis of compound **16n** are as follows: 4-methoxybenzaldehyde (0.1371 g), 4-aminophenol (0.1091 g), benzil (0.2110 g) and ammonium acetate (0.0793 g).

The white solid was obtained with 86% (359 mg) and M.p. = 277-279 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.72 (s, 1H), 7.48 (d, J = 8.5 Hz, 2H), 7.36 (dd, J = 7.0, 2.5 Hz, 2H), 7.31-7.28 (m, 3H), 7.24-7.20 (m, 4H), 7.15 (t, J = 7.5 Hz, 1H), 7.03 (d, J = 8.5 Hz, 2H), 6.83 (dd, J = 6.5, 2.0 Hz, 2H), 6.67 (dd, J = 6.5, 2.5 Hz, 2H), 3.73 (s, 3H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 159.1, 157.2, 146.1, 136.3, 134.7, 131.1, 130.7, 129.8, 129.5, 128.3, 128.1, 128.0, 128.0, 126.3, 126.2, 123.0, 115.6, 113.6, 55.1 ppm.

2-Cyclohexyl-1-(4-hydroxyphenyl)-4,5-triphenyl-1*H* imidazole (16o)



The actual weighed amounts used in the synthesis of compound 16o are as follows: cyclohexanecarbadehyde (0.1123 g), 4-aminophenol (0.112 g), benzil (0.2178 g) and ammonium acetate (0.0789 g).

The white with a slight green hue solid was obtained with 45% (178 mg) and M.p. = 301-304 ^{o}C.

^{1}H NMR (500 MHz, DMSO-d_6): δ = 9.73 (s, 1H), 7.42 (d, J = 7.5 Hz, 2H), 7.27-7.23 (m, 3H), 7.31-7.28 (m, 3H), 7.19 (t, J = 7.5 Hz, 2H), 7.16-7.14 (m, 2H), 7.12-7.09 (m, 1H), 7.06 (dd, J = 6.5, 2.5 Hz, 2H), 6.72 (d, J = 8.5 Hz, 2H), 2.40 (tt, J = 12.0, 3.5 Hz, 1H), 1.80 (d, J = 2.5 Hz, 2H), 1.72 (dd, J = 9.5, 3.5 Hz, 2H), 1.67-1.59 (m, 3H), 1.21 (tt, J = 12.5, 3.5 Hz, 1H), 1.12 (tt, J = 12.5, 3.5 Hz, 2H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 157.2, 152.4, 135.1, 135.0, 131.1, 130.8, 129.5, 129.0, 128.3, 127.9, 127.8, 127.0, 126.2, 125.9, 115.5, 35.4, 31.7, 25.7, 25.4 ppm.

DES [Bet]$_2$[PTSA]

^{1}H NMR (500 MHz, DMSO-d_6): δ = 7.55 (d, J = 7.0 Hz, 2H), 7.21 (d, J = 6.5 Hz, 2H), 4.11 (d, J = 2.0 Hz, 4H), 3.14 (d, J = 2.5 Hz, 18H), 2.23 (d, J = 2.5, 3H) ppm.

^{13}C NMR (125 MHz, DMSO-d_6): δ = 166.6, 142.3, 139.7, 129.4, 125.3, 63.3, 53.9, 53.8, 20.5, 20.4, 20.4 ppm.

Section S5. ^{1}H, and ^{13}C NMR spectrum

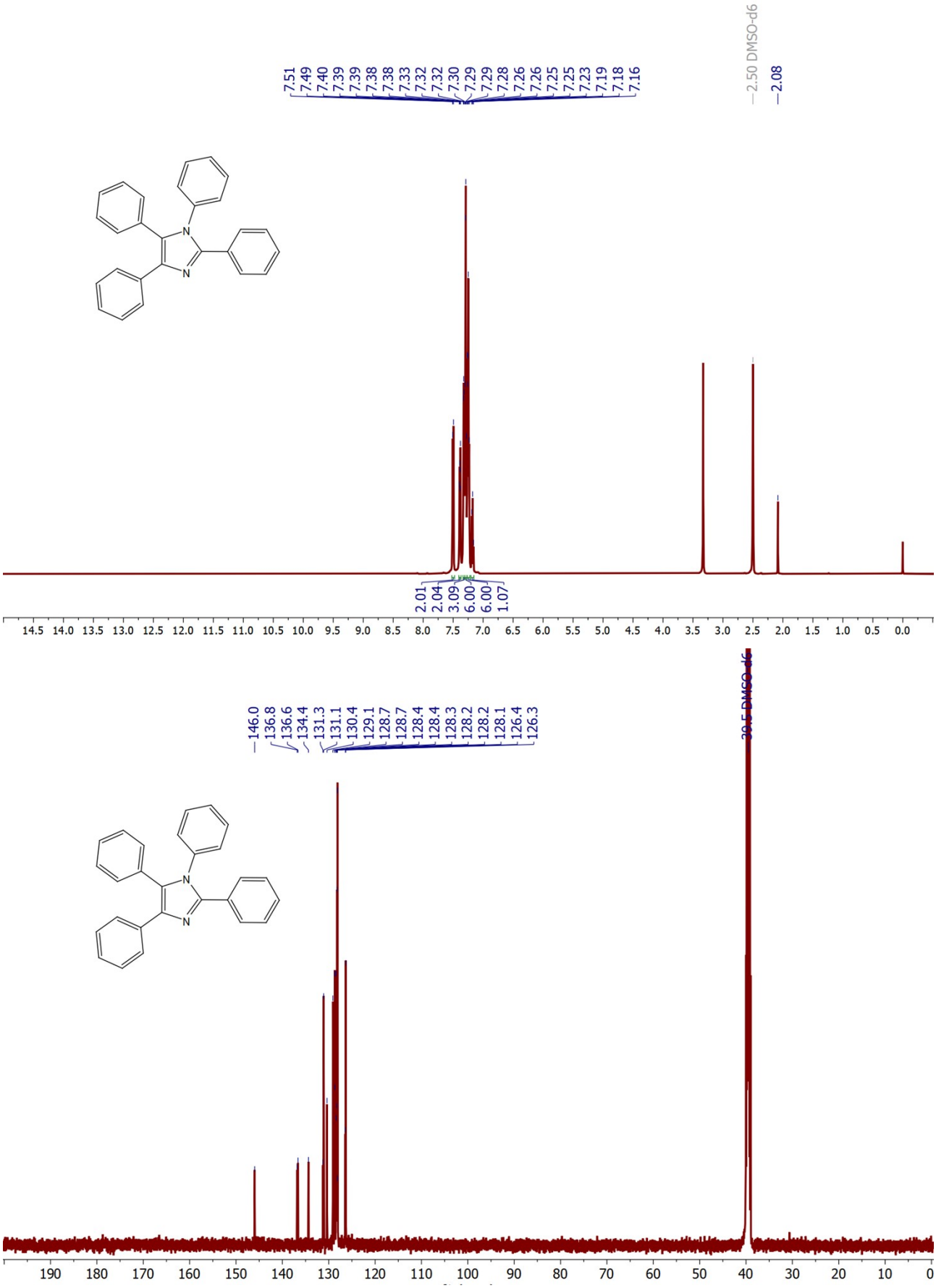


Fig. S4. ^{1}H and ^{13}C -NMR 1,2,4,5-tetraphenyl-1*H*-imidazole (**16a**).

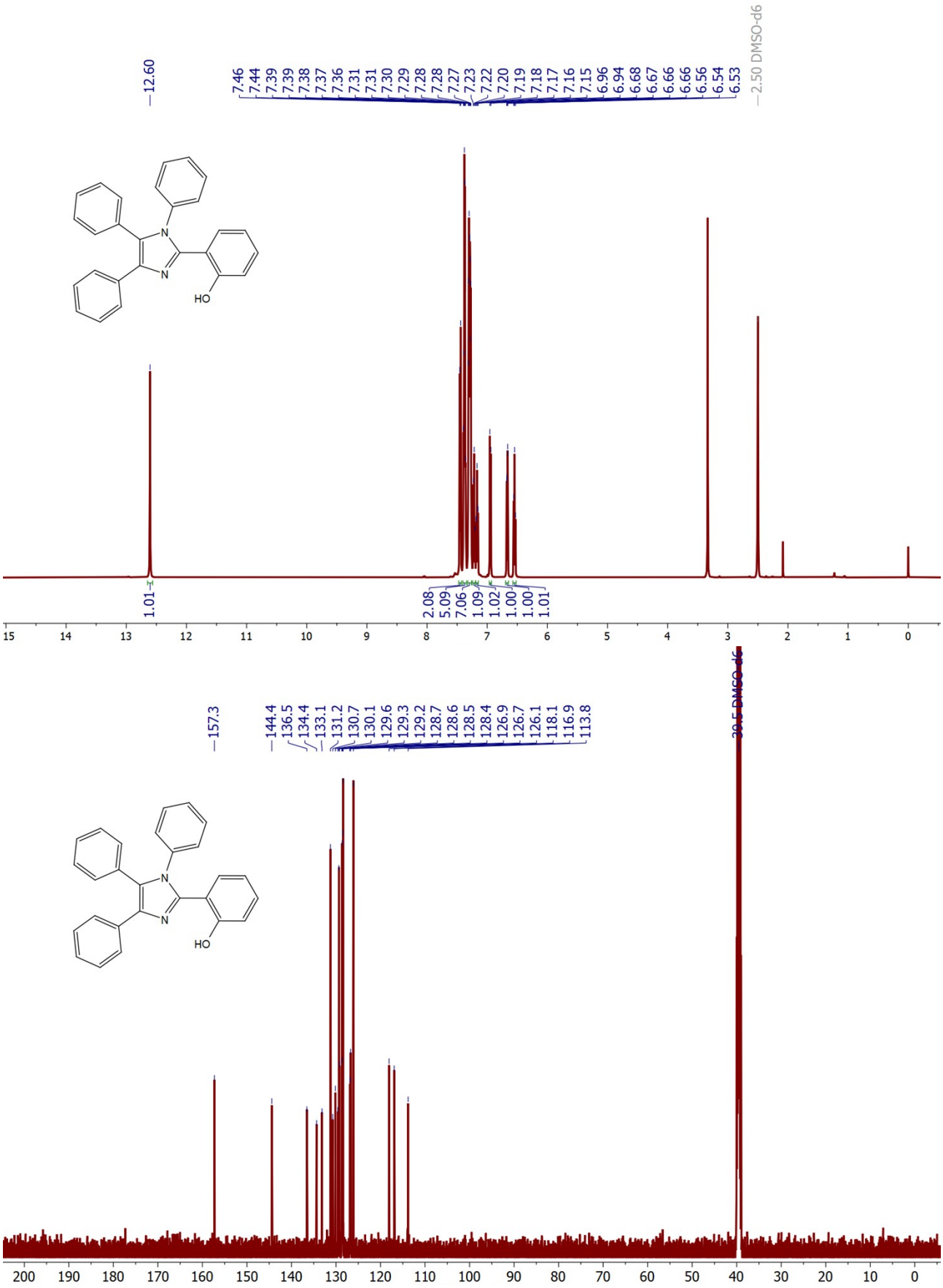


Fig. S5. ^{1}H and ^{13}C-NMR 2-(2-Hydroxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (**16b**).

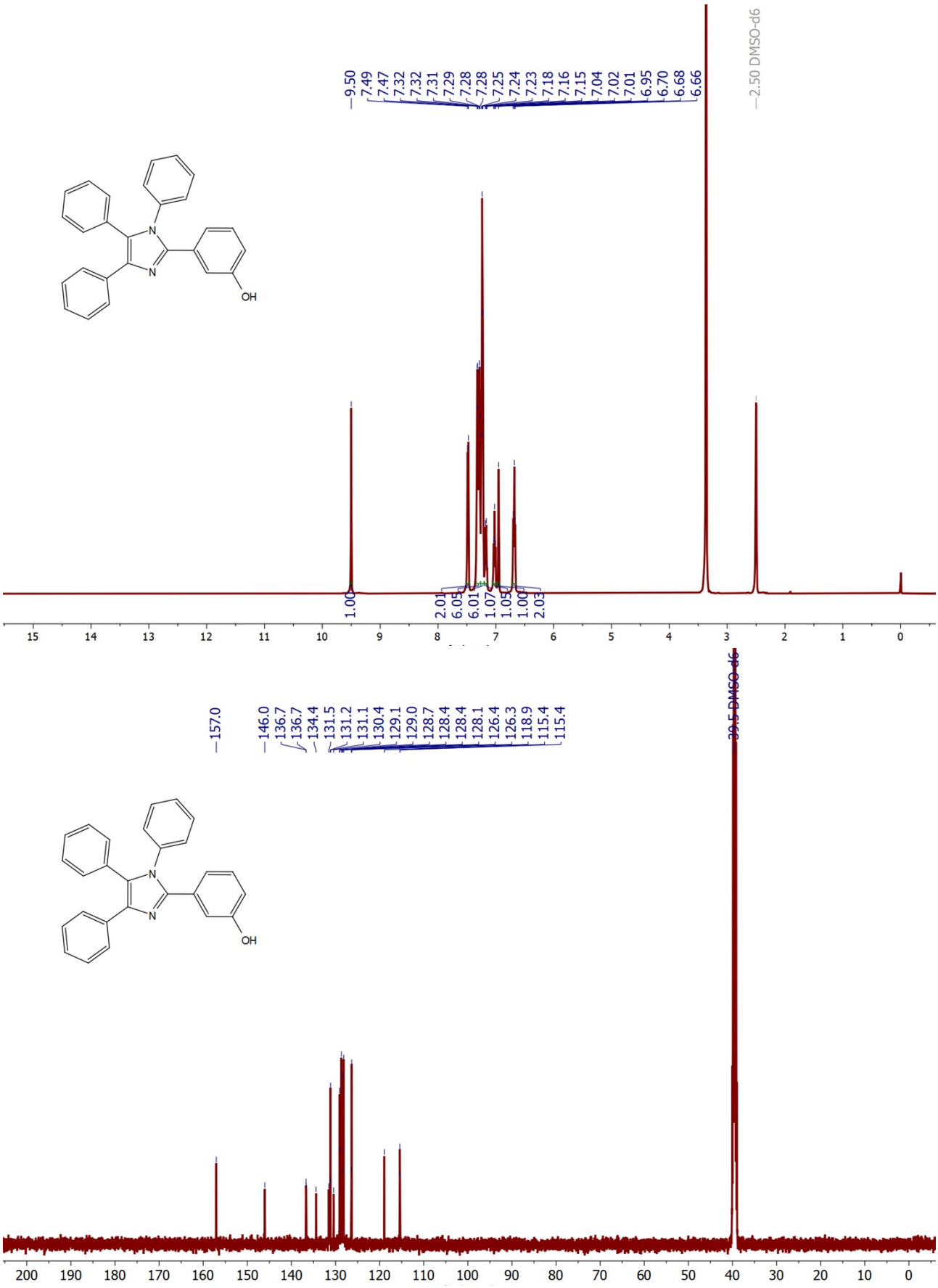


Fig. S6. ^{1}H and ^{13}C-NMR 2-(3-Hydroxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (**16c**).

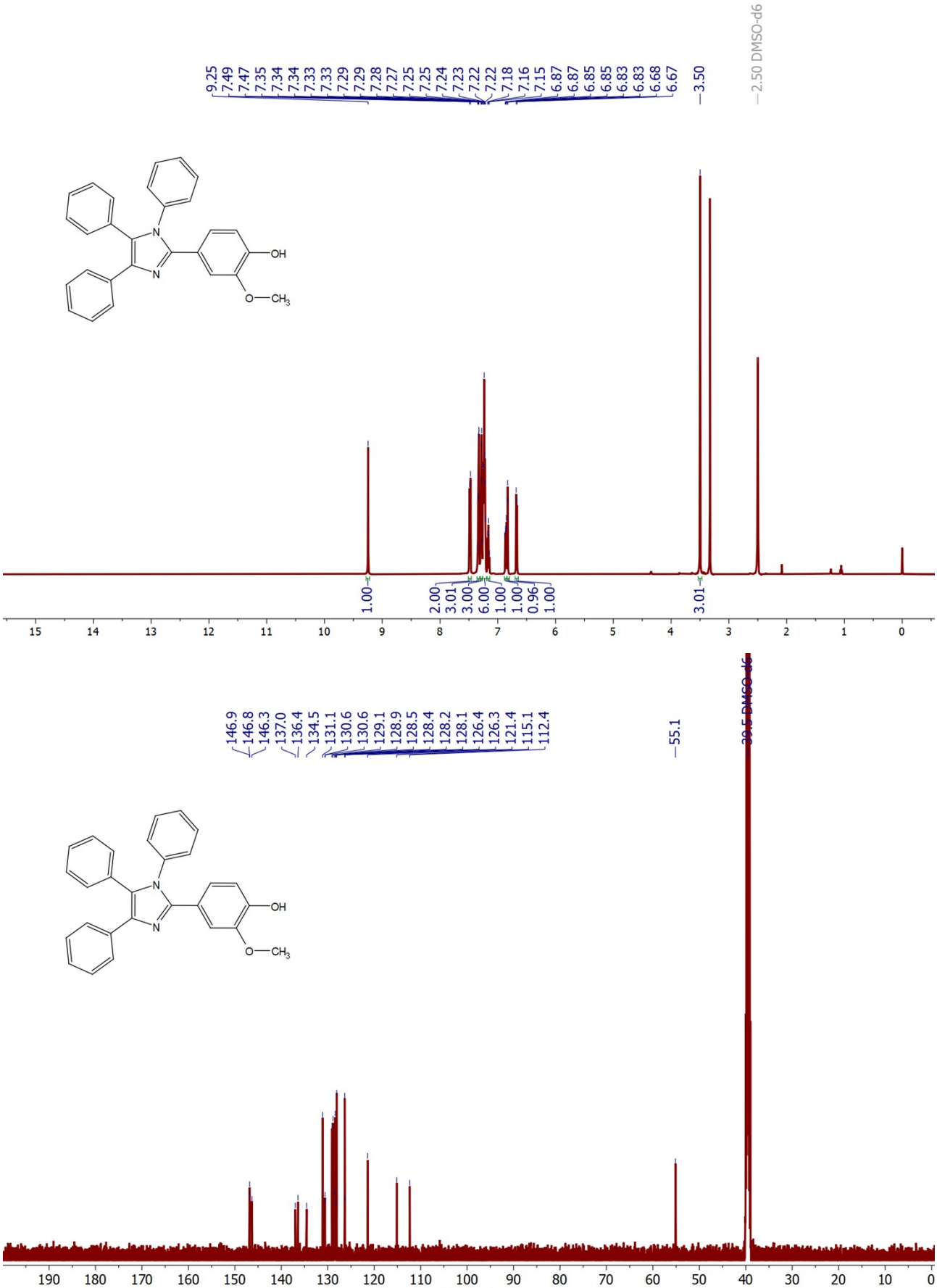


Fig. S7. ^{1}H and ^{13}C-NMR 2-(4-Hydroxy-3-methoxyphenyl)-1,4,5-triphenyl-1*H*-imidazole (**16d**).

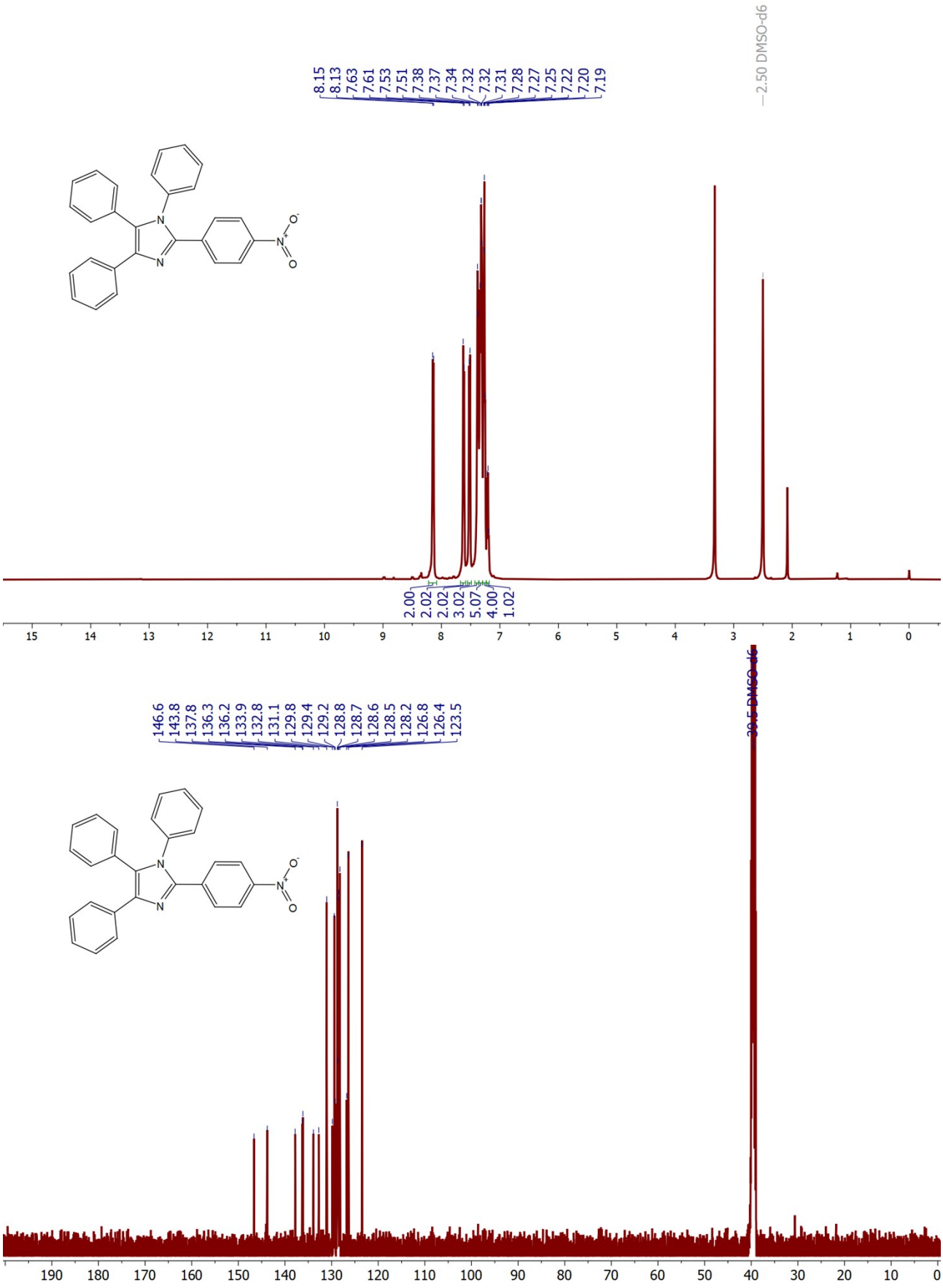


Fig. S8. ^{1}H and ^{13}C-NMR 2-(4-Nitrophenyl)-1,4,5-triphenyl-1*H*-imidazole (**16e**).

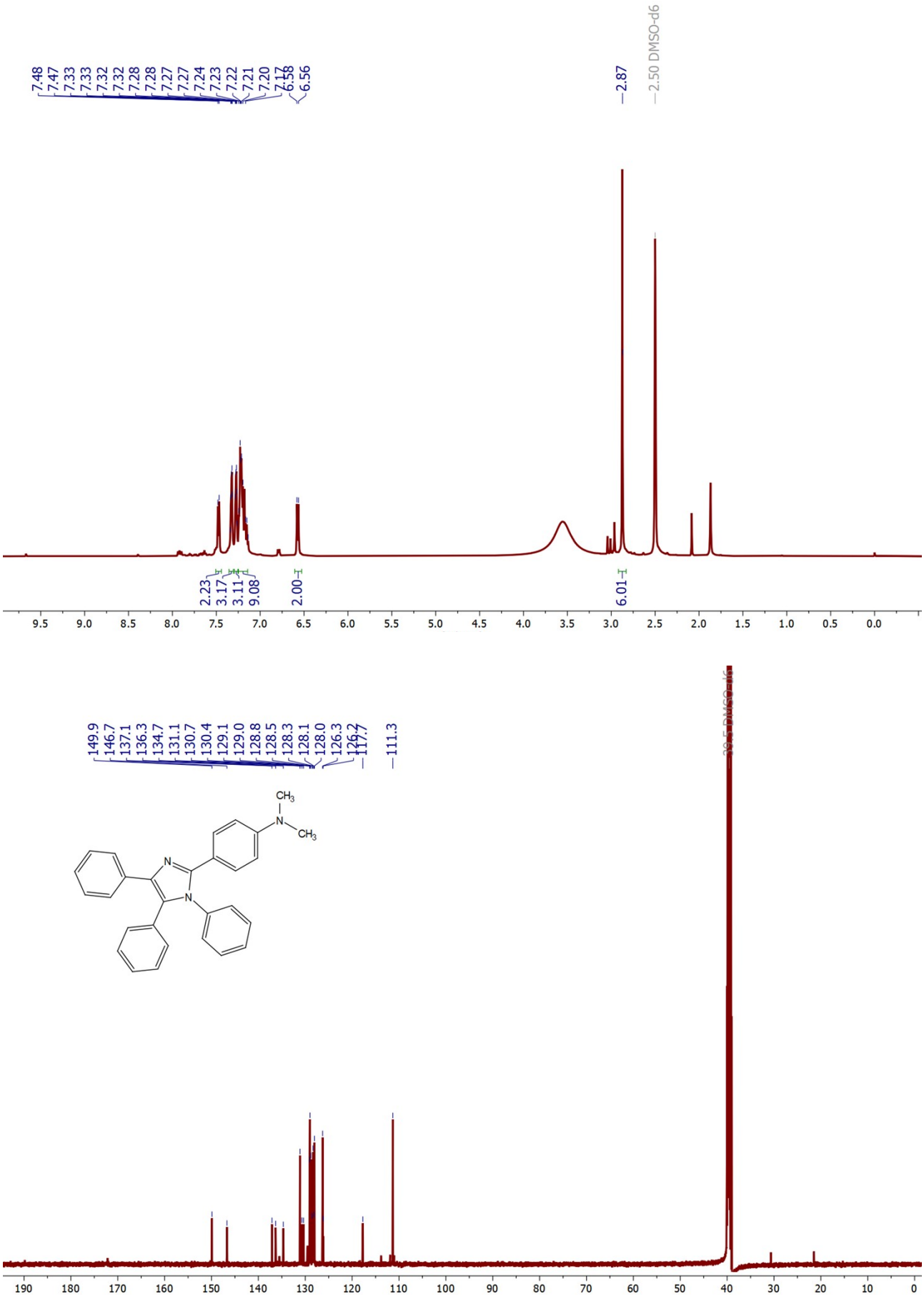


Fig. S9. ^{1}H and ^{13}C-NMR N,N-dimethyl-4-(1,4,5-triphenyl-1H-imidazol-2-yl)aniline (**16f**).

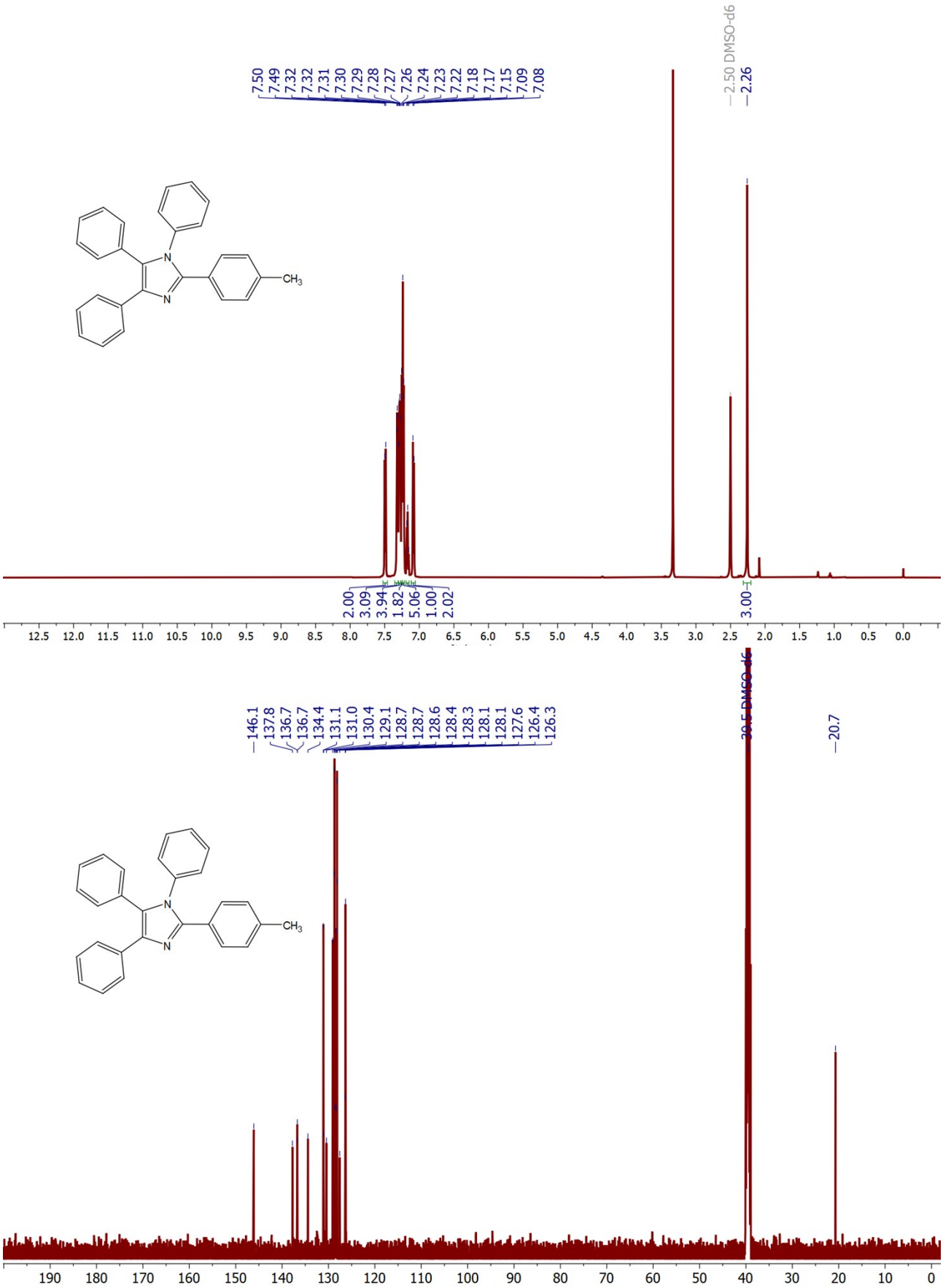


Fig. S10. ^{1}H and ^{13}C-NMR 2-(4-Methylphenyl)-1,4,5-triphenyl-1*H*-imidazole (**16g**).

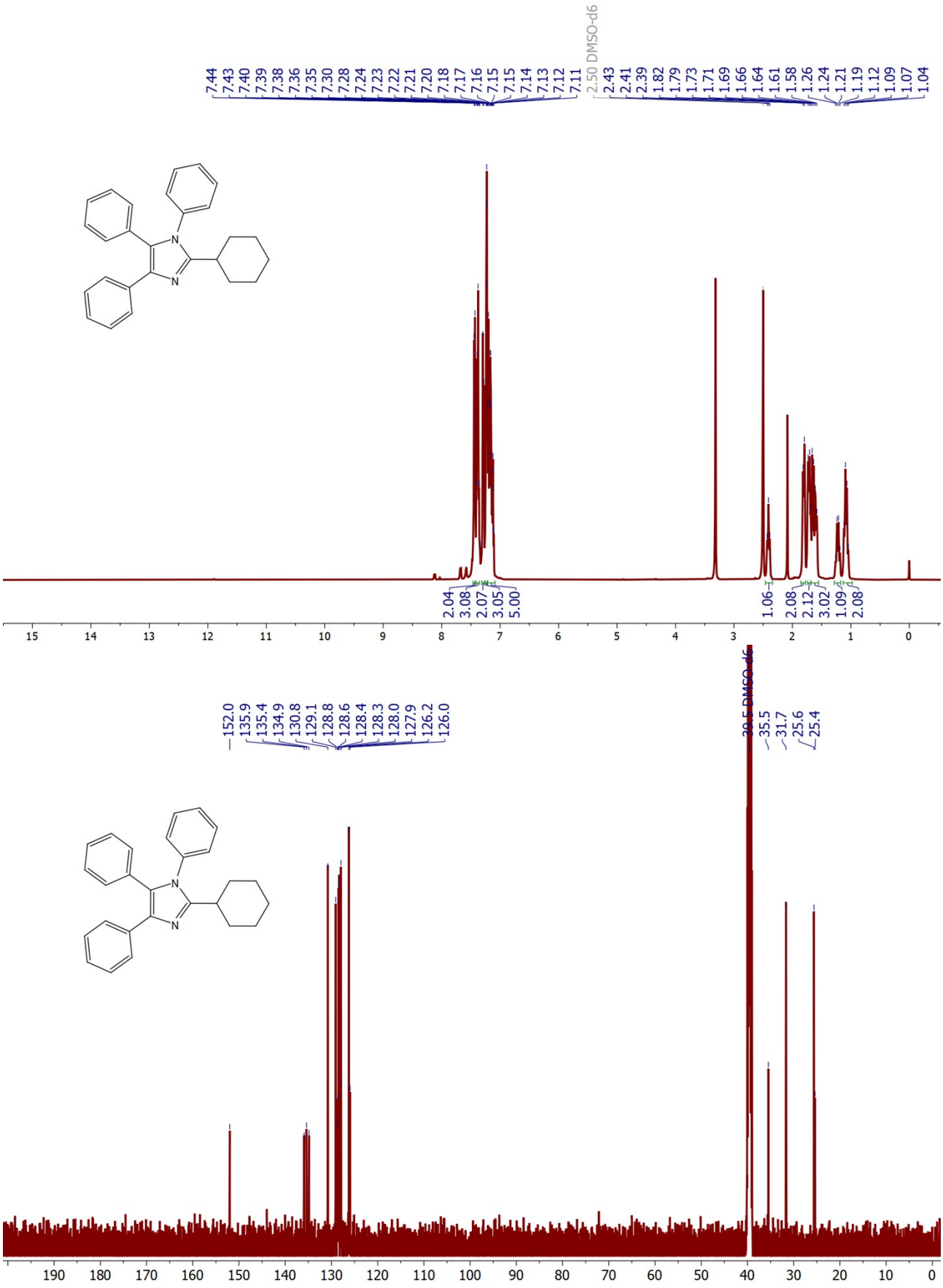


Fig. S11. ^{1}H and ^{13}C-NMR 2-Cyclohexyl-1,4,5-triphenyl-1*H*-imidazole (**16h**).

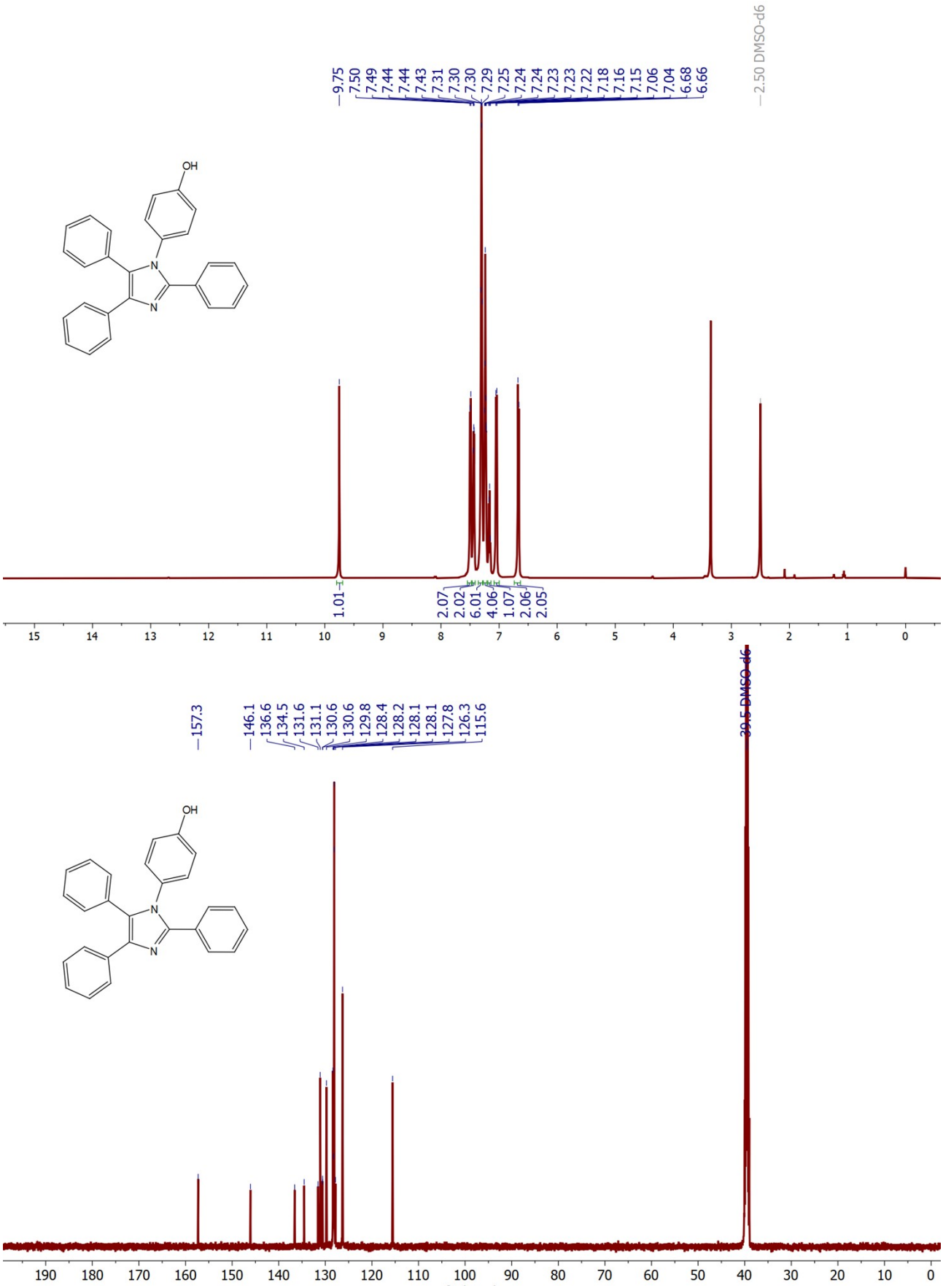


Fig. S12. ^{1}H and ^{13}C-NMR 1-(4-Hydroxyphenyl)-2,4,5-triphenyl-1*H*-imidazole (**16i**).

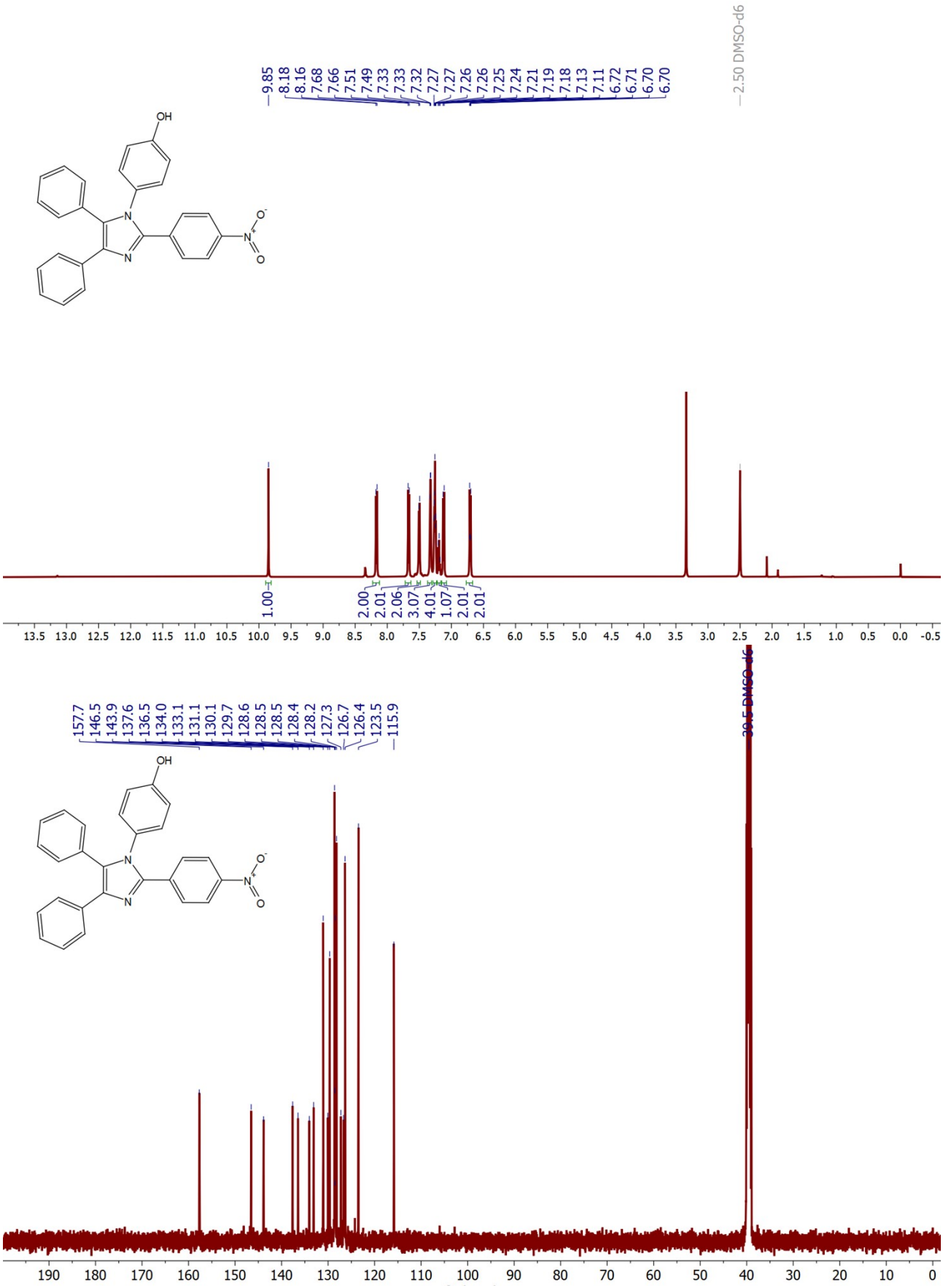


Fig. S13. ^{1}H and ^{13}C-NMR 1-(4-Hydroxyphenyl)-2-(4-nitrophenyl)-4,5-diphenyl-1*H*-imidazole (**16j**).

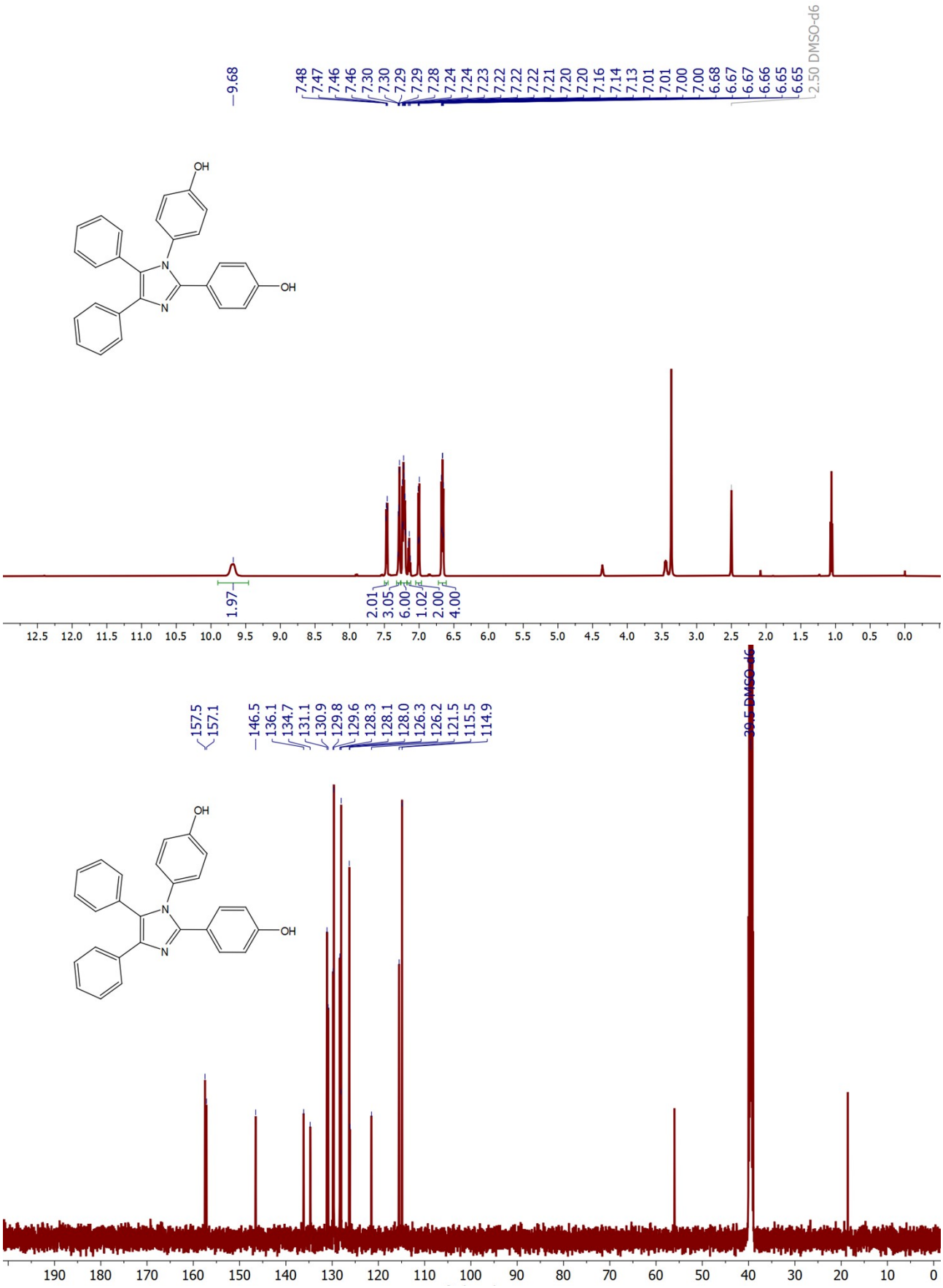


Fig. S14. ^{1}H and ^{13}C-NMR 1,2-(4-Hydroxyphenyl)-4,5-diphenyl-1*H*-imidazole (**16k**).

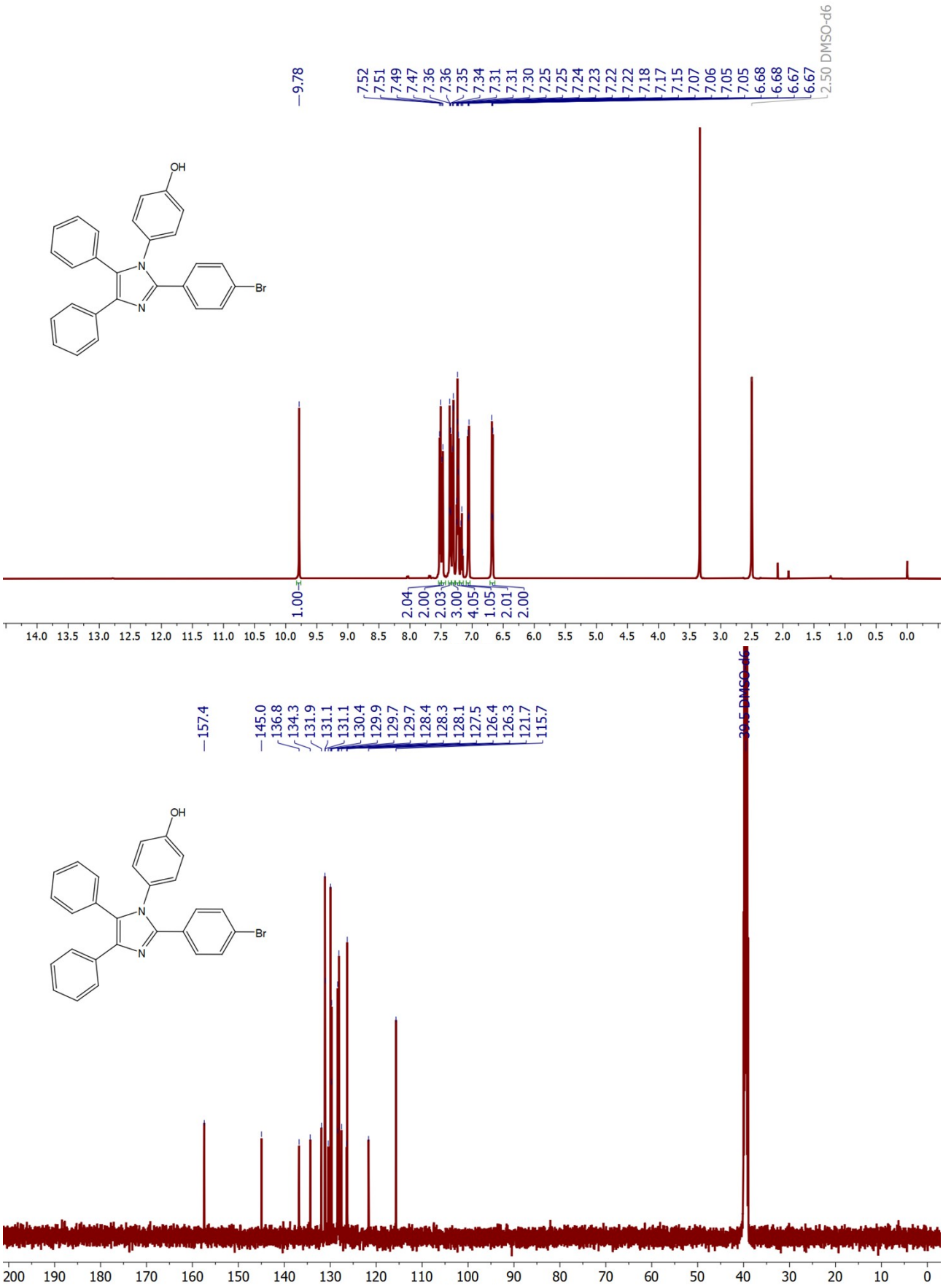


Fig. S15. ^{1}H and ^{13}C-NMR 1-(4-Hydroxyphenyl)-2-(4-bromophenyl)-4,5-diphenyl-1*H*-imidazole (**16l**).

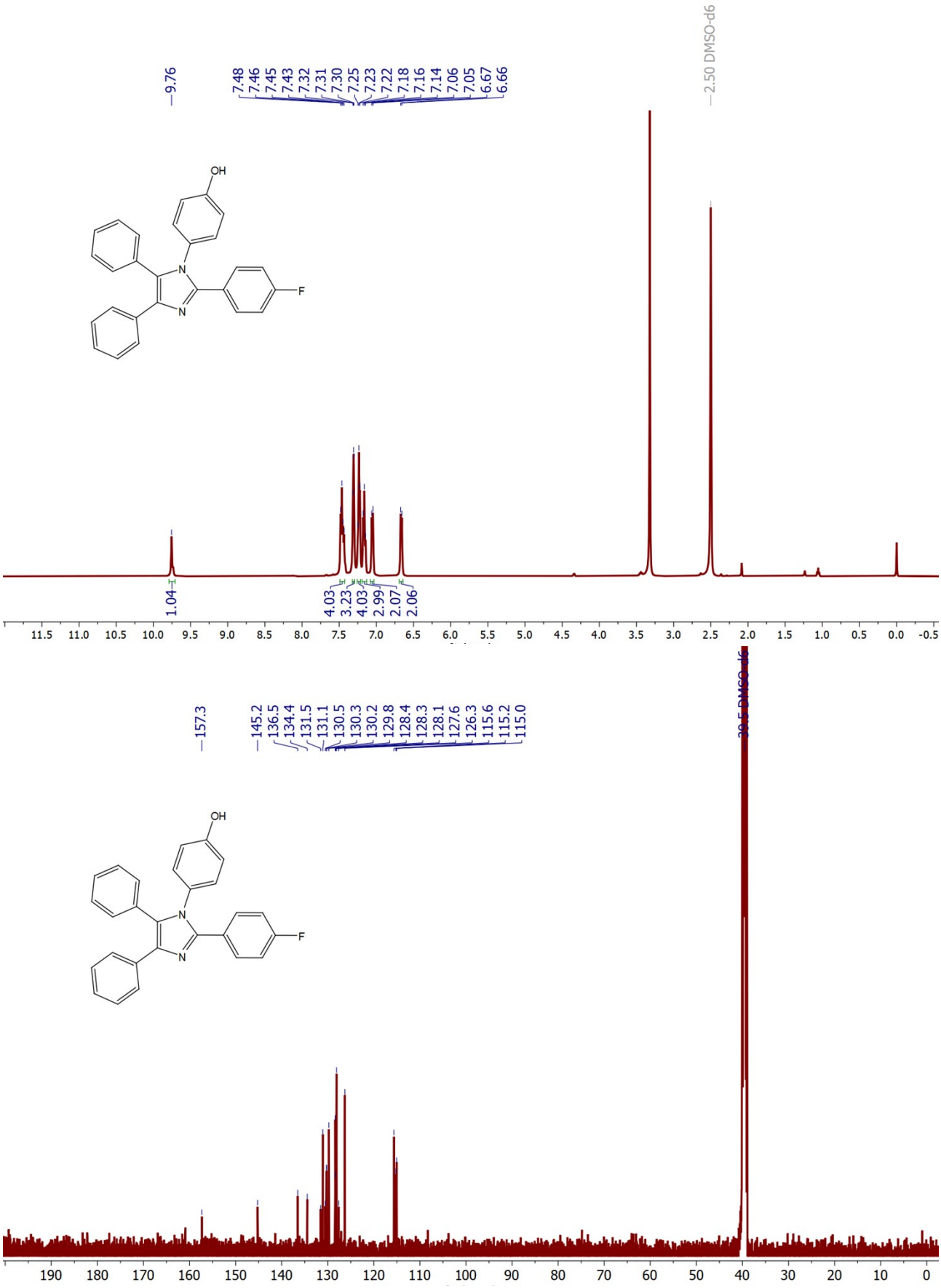


Fig. S16. ^{1}H and ^{13}C-NMR 1-(4-Hydroxyphenyl)-2-(4-flourophenyl)-4,5-diphenyl-1H-imidazole (**16m**).

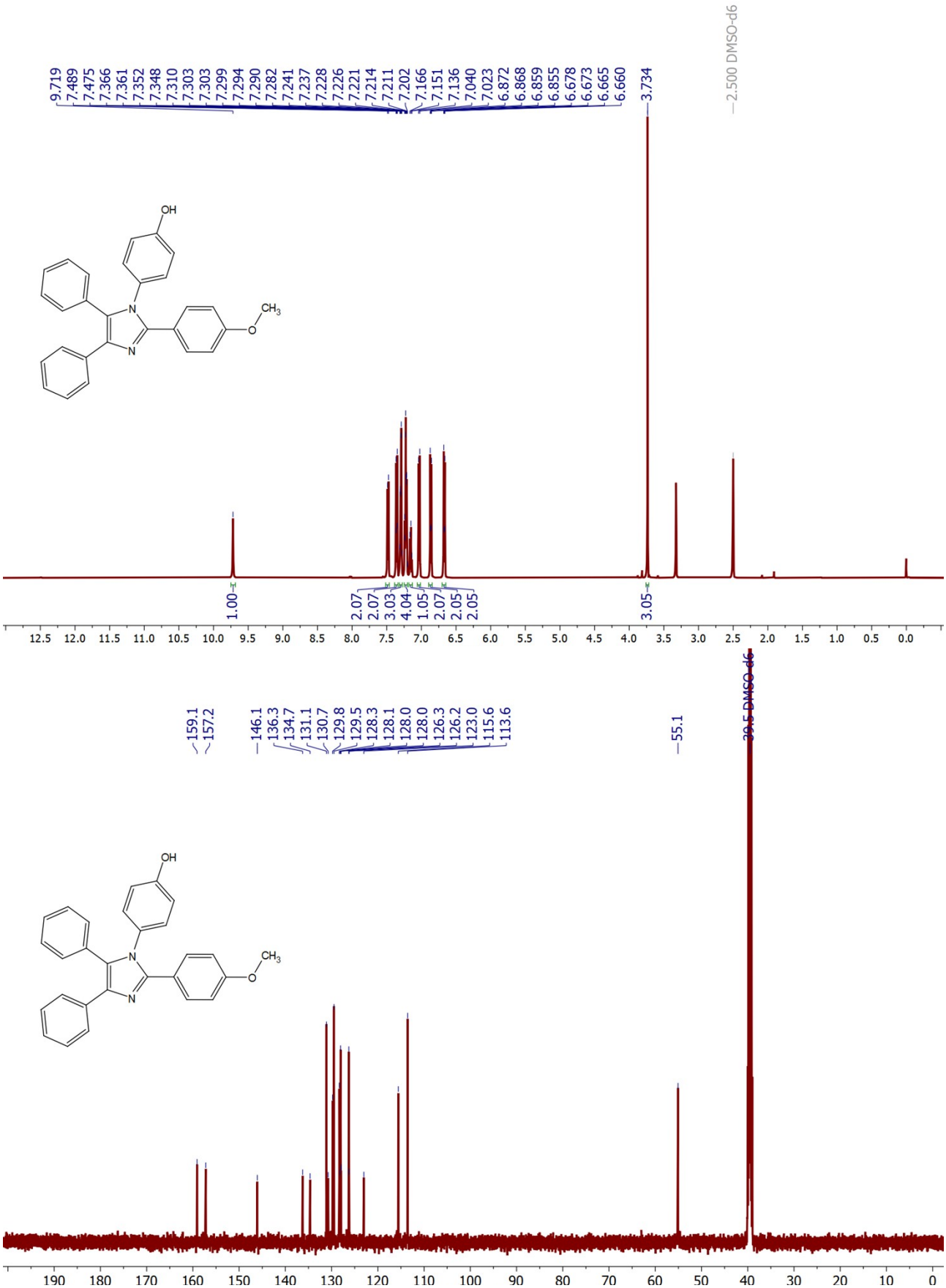


Fig. S17. ^{1}H and ^{13}C-NMR 1-(4-Hydroxyphenyl)-2-(4-methoxyphenyl)-4,5-diphenyl-1*H*-imidazole (**16n**).

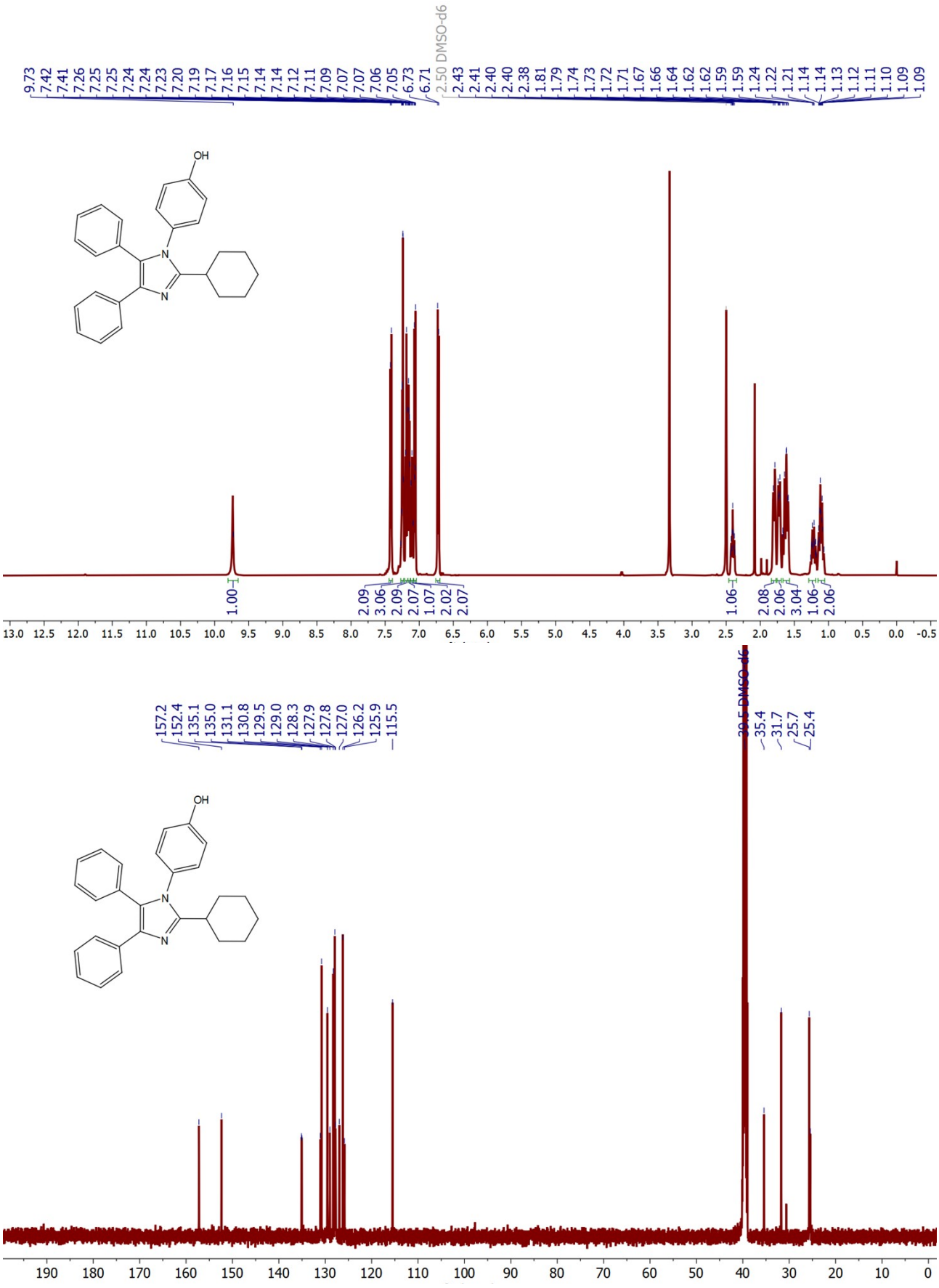


Fig. S18. ^{1}H and ^{13}C-NMR 2-Cyclohexyl-1-(4-hydroxyphenyl)-4,5-triphenyl-1*H* imidazole (**16o**).

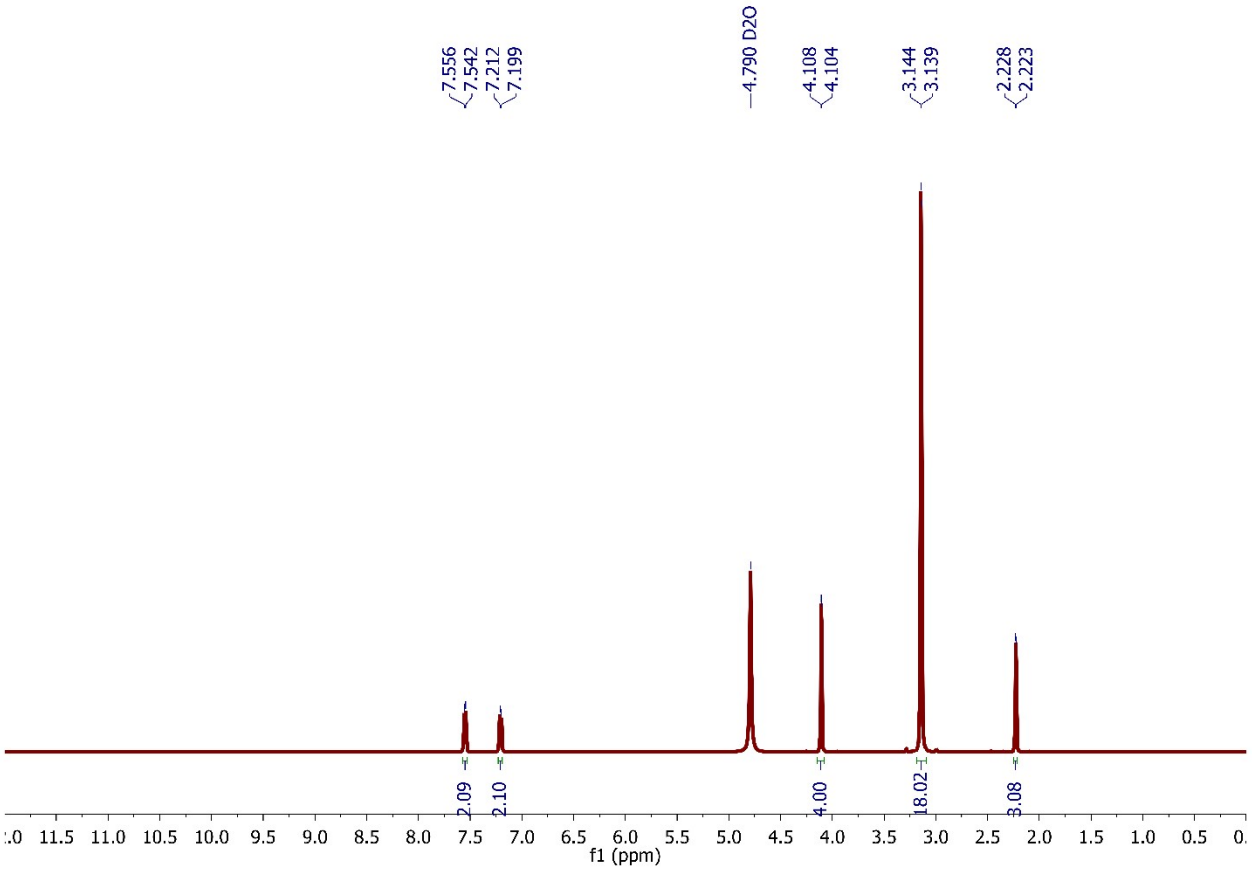


Fig. S19. ^{1}H-NMR DES $[Bet]_2[PTSA]$.

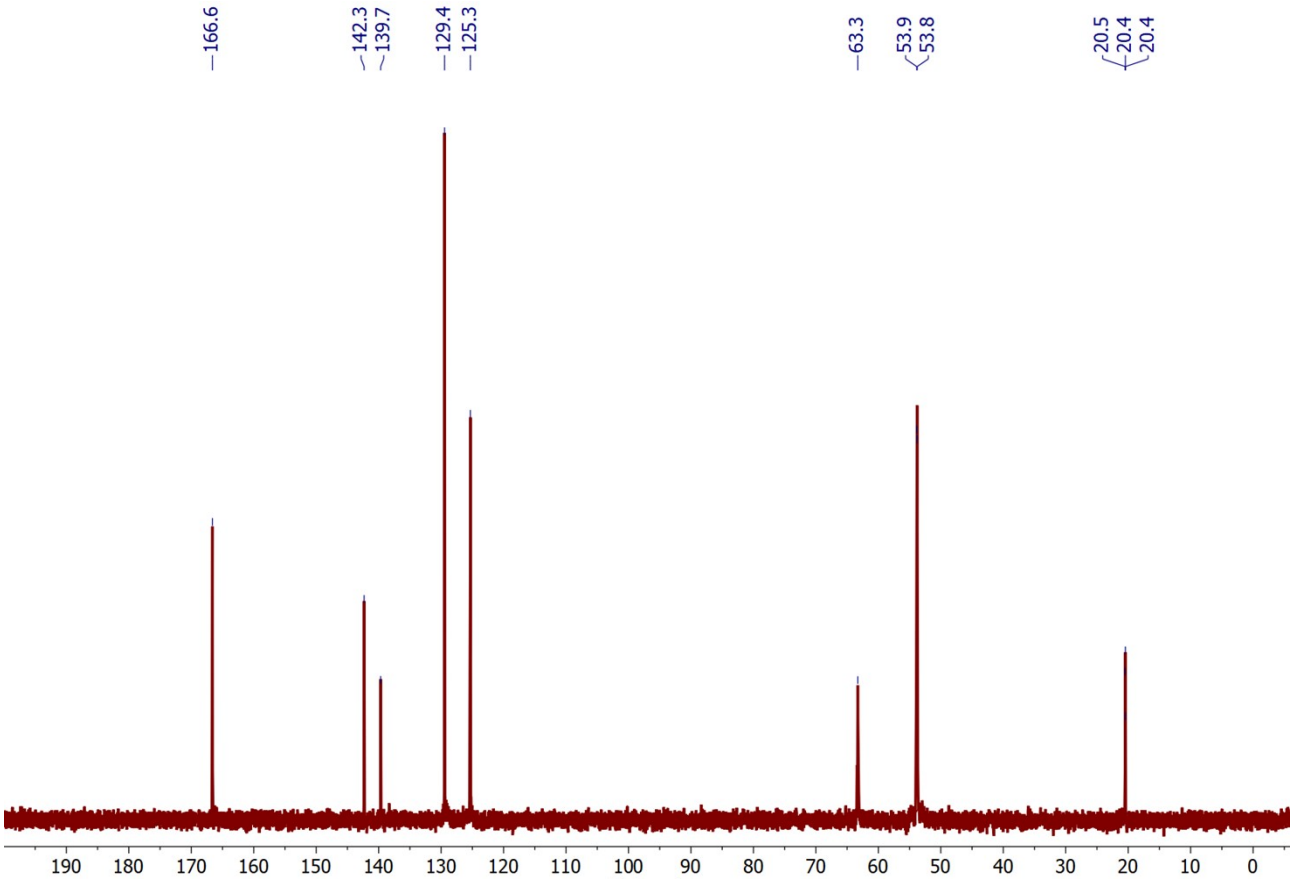


Fig. S20. ^{13}C-NMR DES $[Bet]_2[PTSA]$.

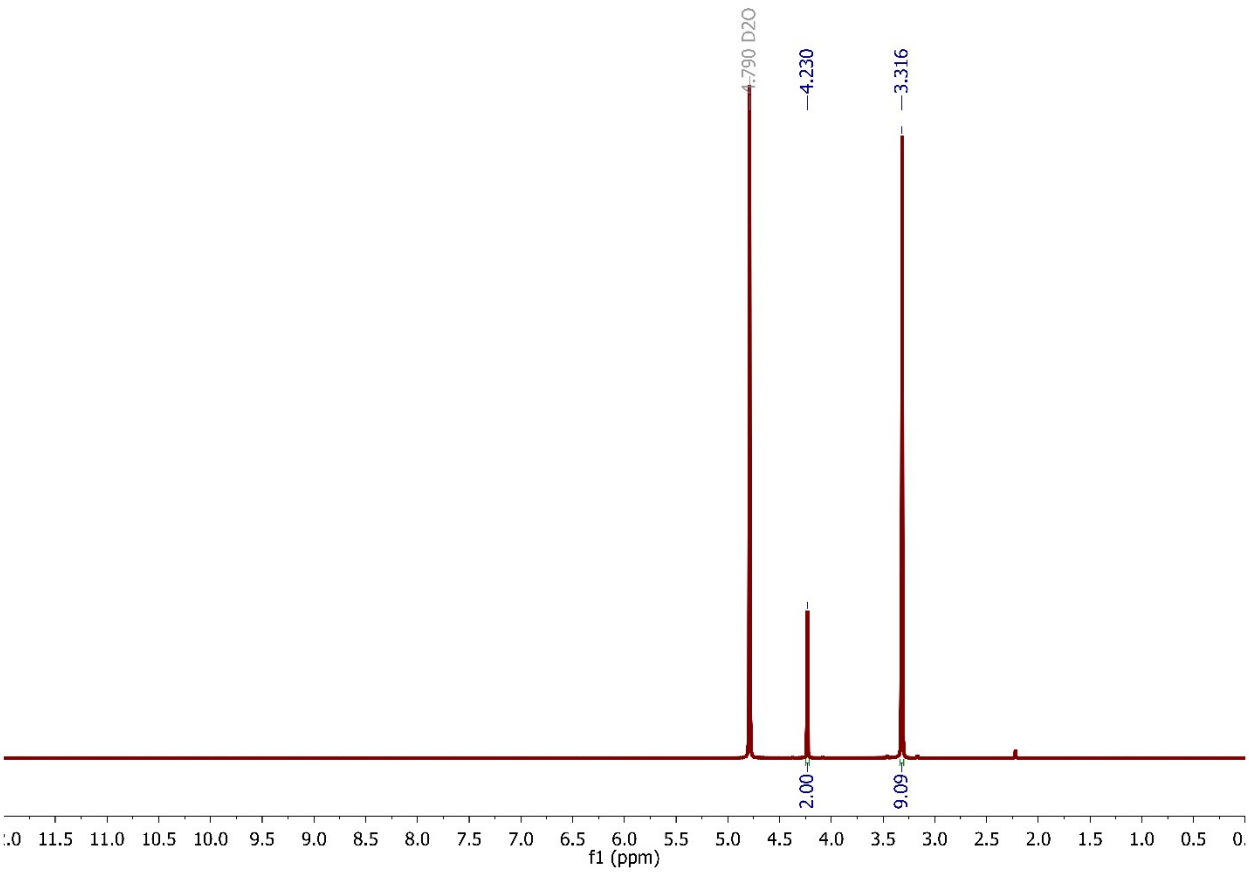


Fig. S21. ^{1}H-NMR of Betaine.

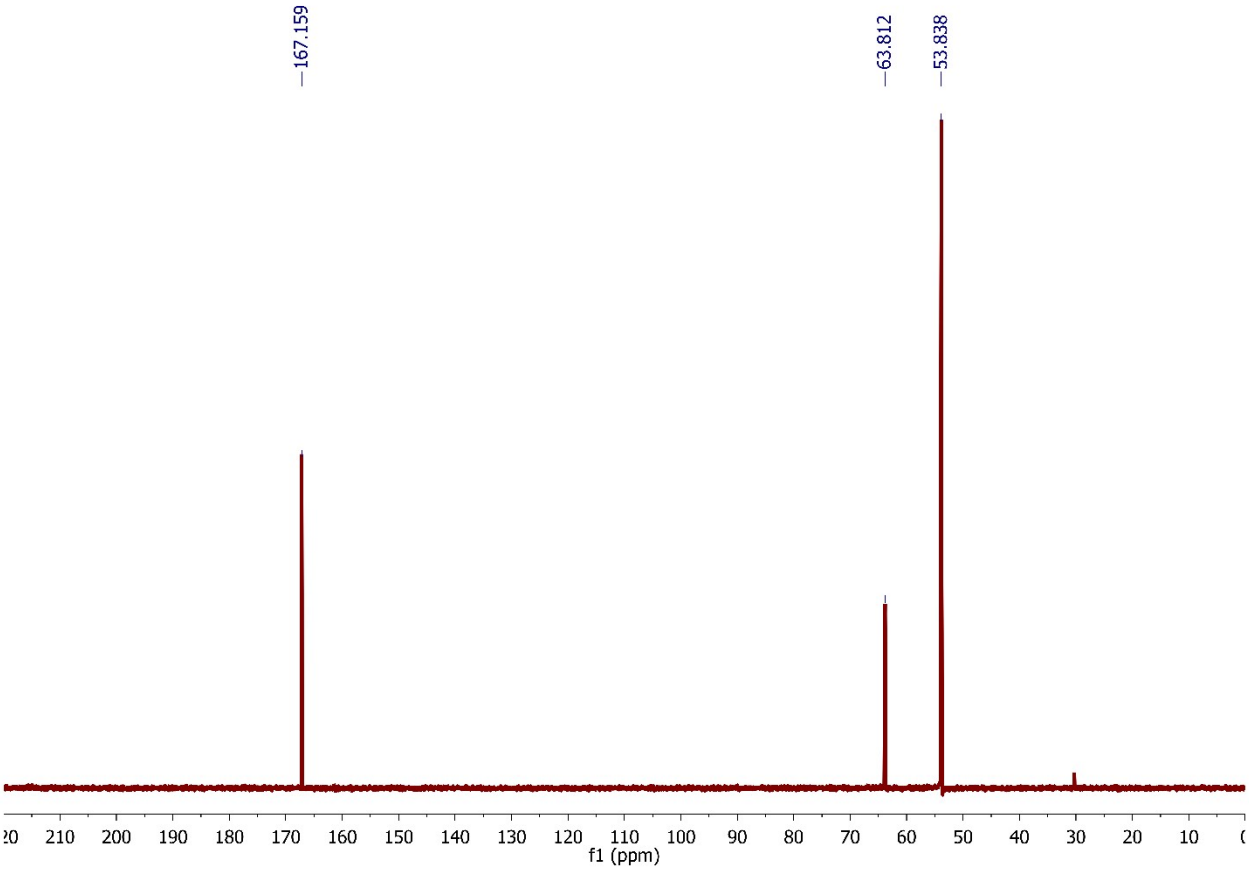


Fig. S22. ^{13}C-NMR of Betanine.

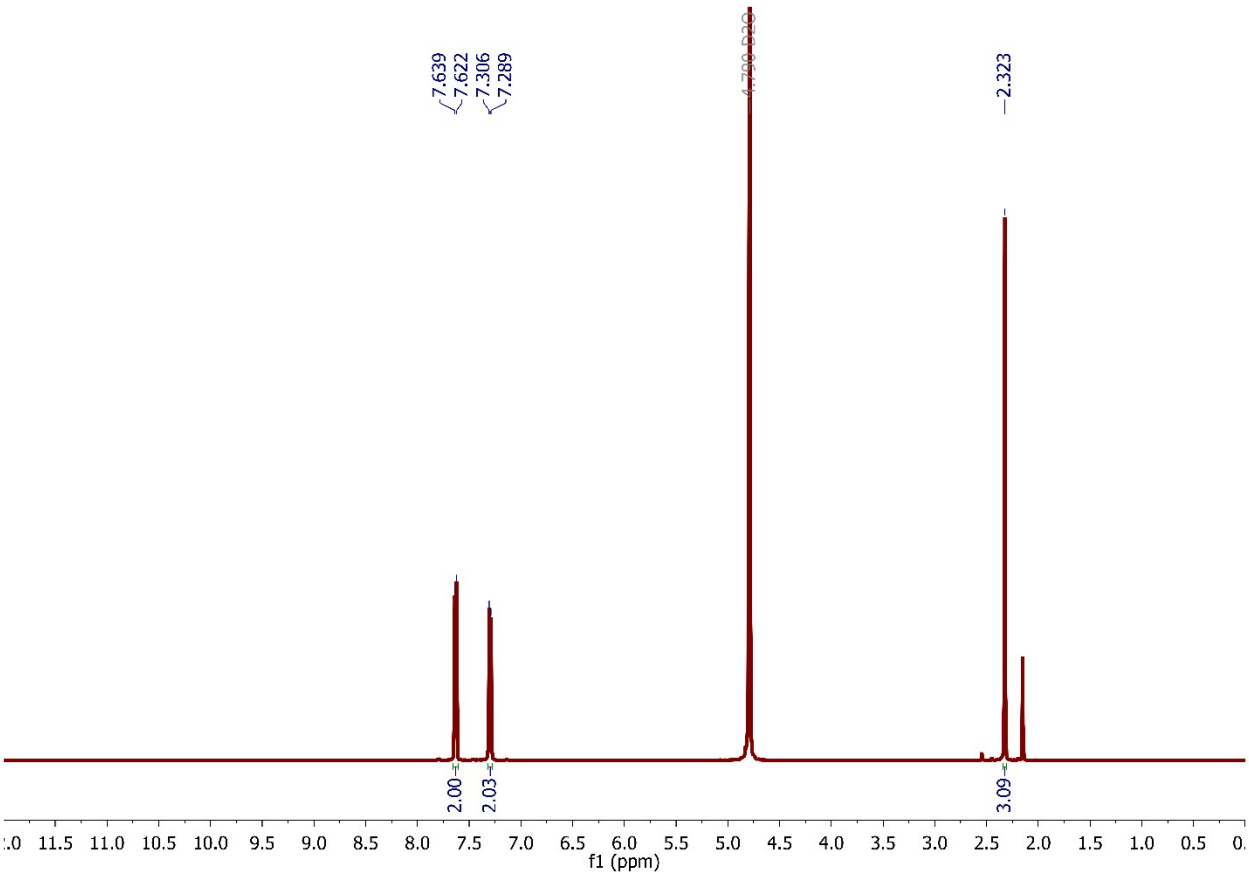


Fig. S23. ^{1}H-NMR of PTSA.

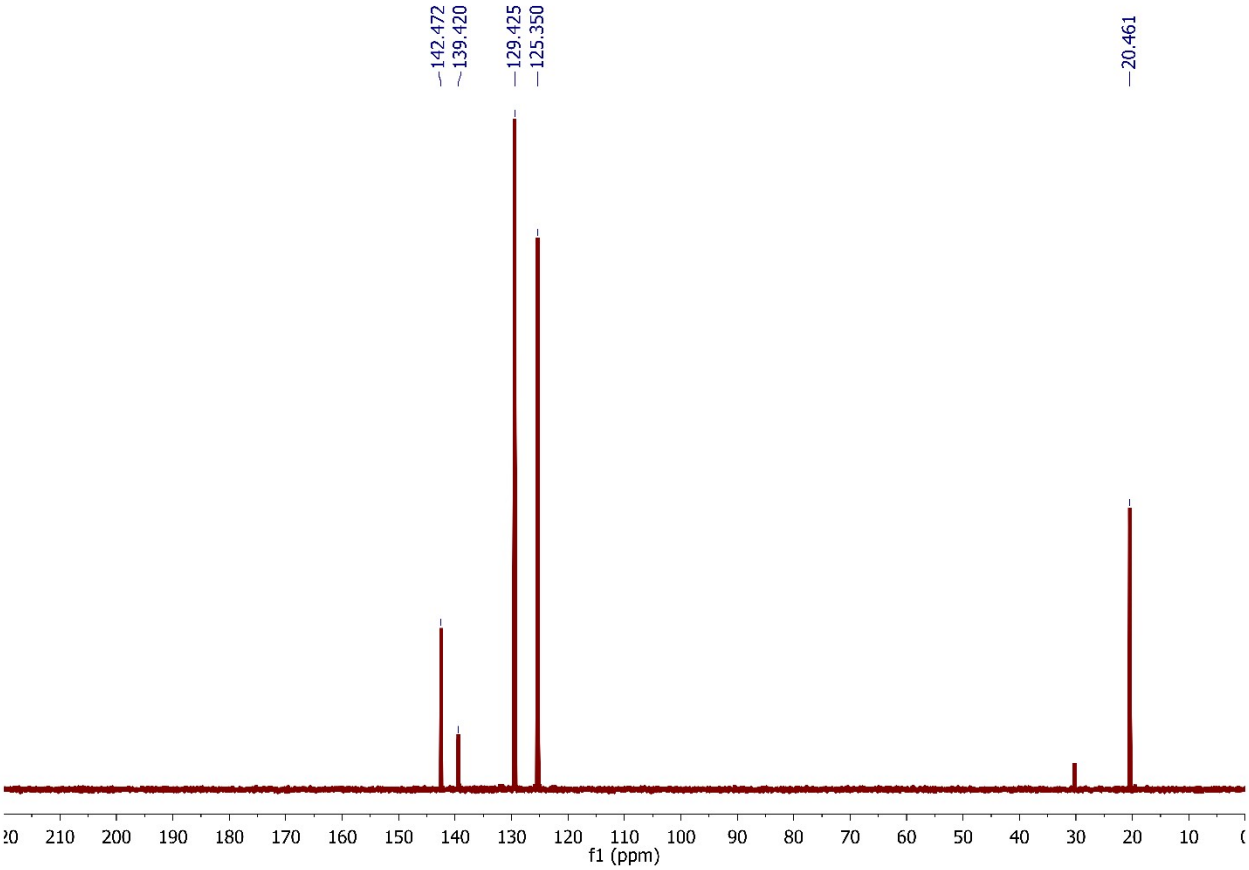


Fig. S24. ^{13}C-NMR of PTSA.