Electronic Supplementary Information

A Double Chain Based Metallomicellar Catalyst for Aerobic Oxidative Synthesis of Benzimidazoles in Water

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S1. Materials and methods

3-Aminophenol, bis(acetylacetonato)dioxomolybdenum (VI), 1-bromohexadecane, bromomethane, 2,4-dihydroxybenzaldehyde were purchased from Sigma-Aldrich. Ethanolamine, $POCl_3$, ^{i}PrOH, and ethyl acetate were purchased from SISCO research laboratories Pvt. Ltd., India. The benzylamines **1a-3a**, **6a-8a**, **11a**, **13a, 15a,** and the deuterated solvents were purchased from Sigma-Aldrich, whereas **4a, 5a, 16a,** and **17a** were purchased from TCI Chemicals (India Pvt. Ltd.). Benzylamines **9a, 10a, 12a, 14a,** and **19a** were purchased from BLD Chemicals and the 1,2-diaminobenzenes, **1b** and **2b** were purchased from Sigma-Ardrich. All the aforementioned purchased chemicals from different sources were used without any further purification.

The NMR spectra were recorded on a Bruker Avance-400 and 500 instruments. Fourier Transform Infrared (FT-IR) spectra of the complexes were measured on a model Nexus 670 (FTIR), Centaurms 10X (Microscope) having spectral Range 4,000 to 400 cm^{-1} with a MCT-B detector. ESI-Mass spectrum of the complex was recorded on Agilent Q-TOF spectrometer in a positive ion mode. HR-TEM images were obtained on HR-TEM, (TecnaiTM G2 TF20) working at an accelerating voltage of 200 kV. The wavelength of the laser used was 632.8 nm with the scattering angle of 90°. LMCM-20 conductivity meter from LABMAN scientific instruments was used for the determination of critical micellar concentration of the complexes. DFT study was performed with the Gaussian 09 software package. The B3LYP (Becke's three parameter hybrid functional using the LYP correlation) functional was used for geometry optimizations and frequencies with LANL2DZ for Mo atom, and the 6-31G* basis set for carbon, nitrogen, oxygen and hydrogen. Frequency calculations were performed for the optimized structures to confirm the absence of any imaginary frequencies.

S2. Synthesis and analytical data of the oxomolybdenum complexes $MoO_2L_1(OH_2)$, Mo1 and $MoO_2L_2(OH_2)$, Mo2

The reproduction of oxomolybdenum complexes **Mo1** and **Mo2** was carried out following the earlier report of our laboratory.[1] In this line, the respective ligand, $\mathbf{L_1}(H)_2$ or $\mathbf{L_2}(H)_2$ (1.0 mmol) was added to a solution of bis(acetylacetonato)dioxomolybdenum(VI) (1.0 mmol) in 20 mL methanol. The mixture was stirred for 3 hours under reflux condition and a clear yellow-colored solution was observed. After completion of the reaction, the solution was cooled down and allowed to stand overnight to obtain yellow-colored semi-crystalline solid. The solid was filtered out and air-dried. The obtained solid was further stirred in water (20 mL) at room temperature for 1 hour. The precipitate was separated by filtration, washed with cold water, and dried under vacuum to obtain the desired complexes **Mo1** and **Mo2**.

(1) $MoO_2L_1(OH_2)$, Mo1

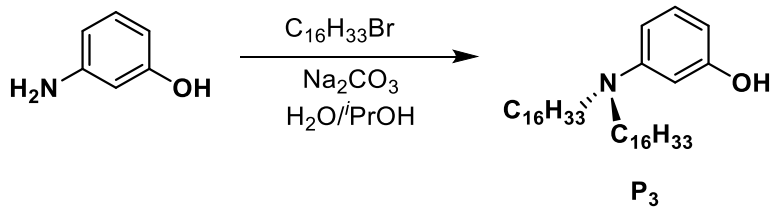
^{1}H NMR (DMSO-d_6, 400 MHz, 24 °C): δ 8.61 (s, 1H), 7.46-7.44 (m, 1H), 6.56-6.53 (m, 1H), 6.44 (s, 1H), 4.41-4.38 (m, 2H), 3.98-3.95 (m, 2H), 3.79-3.78 (m, 3H) *ppm*.

^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 164.75, 163.27, 162.83, 134.90, 114.70, 107.45, 102.96, 71.50, 60.82, 55.45 *ppm*.

(2) $MoO_2L_2(OH_2)$, Mo2

^{1}H NMR (DMSO-d_6, 500 MHz, 100 °C): δ 8.56 (s, 1H), 7.42-7.40 (m, 1H), 6.53-6.50 (m, 1H), 6.40-6.39 (m, 1H), 4.45-4.42 (m, 2H), 4.03-3.98 (m, 4H), 1.74-1.71 (m, 2H), 1.44-1.39 (m, 2H), 1.27 (m, 24 H), 0.89-0.86 (m, 3H) *ppm*.

S3. Synthesis and analytical data of the precursor, 3-(dihexadecyl)aminophenol (P_3)



Scheme 1 Synthesis of precursor, 3-(dihexadecyl)aminophenol.

Precursor 3-(dihexadecyl)aminophenol was synthesized following the literature report[2] with necessary modification taking 3-aminophenol and 1-bromohexadecane as starting material. 3-Aminophenol (1.0 g, 9.16 mmol), 1-bromohexadecane (5.59 g, 18.32 mmol) and Na_2CO_3 (1.94

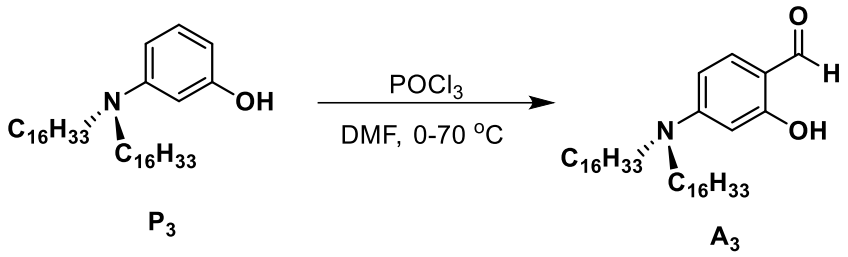
g, 18.32 mmol) were taken in H_2O (10 mL) and ^{i}PrOH (10 mL) solvent mixture. The reaction mixture was allowed to stir for 12 hours under reflux conditions. After completion of the reaction, the solvent was removed under reduced pressure. The obtained crude product was purified by column chromatography using hexane and ethylacetate as eluent.

^{1}H NMR (400 MHz, $CDCl_3$, 24 ^{o}C): δ 7.05-7.01 (m, 1H), 6.24-6.22 (m, 1H), 6.13-6.09 (m, 2H), 3.23-3.19 (m, 4H), 1.56 (m, 4H), 1.30-126 (m, 52H), 0.90-0.87 (m, 6H) *ppm*.

^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$, 24 ^{o}C): δ 156.85, 149.91, 130.16, 104.84, 102.29, 98.73, 51.31, 32.08, 29.85, 29.78, 29.71, 29.52, 27.40, 27.34, 22.84, 14.27 *ppm*.

HRMS (ESI) m/z: calculated for $C_{38}H_{71}NO$+H: 558.5534; found: 558.5609

S4. Synthesis and analytical data of precursor, A_3



Scheme 2 Synthesis of precursor, $\mathbf{A_3}$.

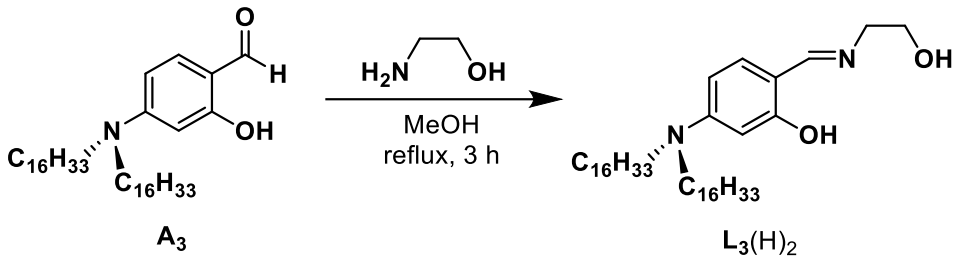
Aldehyde, $\mathbf{A_3}$ was prepared by the Vilsmeier–Haack method following a literature protocol.[3] Initially, 5.0 mL of dry DMF was cooled to 0-5 °C in an ice bath and $POCl_3$ (0.34 g, 2.24 mmol) was added dropwise into it. The solution was heated at 40 °C for half an hour and a yellow-colored Vilsmeier reagent appeared. The solution was again cooled to 0-5 °C and 3-(hexadecyl)aminophenol (0.5 g, 0.89 mmol) was added dropwise with continuous stirring. The reaction mixture was allowed to stir for 15-20 minutes and heated at 70 °C for 6 hours. After completion of the reaction, the ice-cooled distilled water was added to the reaction mixture. The organic layer was separated from the aqueous layer by phase separation by adding DCM. The organic solvent was evaporated under reduced pressure and the desired product $\mathbf{A_3}$ was purified by column chromatography using hexane and ethyl acetate as eluent.

^{1}H NMR (500 MHz, $CDCl_3$, 24 ^{o}C): δ 11.64 (s, 1H), 9.48 (s, 1H), 7.25-7.21 (m, 1H), 6.23-6.21 (m, 1H), 6.03 (s, 1H), 3.31-3.28 (m, 4H), 1.61-1.60 (m, 4H), 1.31-126 (m, 52H), 0.89-0.86 (m, 6H) *ppm*.

$^{13}C\{^{1}H\}$ NMR (126 MHz, $CDCl_3$, 24 °C): δ 191.95, 164.44, 154.68, 135.38, 111.51, 104.69, 96.99, 51.38, 32.07, 29.84, 29.80, 29.78, 29.74, 29.71, 29.58, 29.50, 27.43, 27.16, 22.83, 14.25 *ppm*.

HRMS (ESI) m/z: calculated for $C_{39}H_{71}NO_2$+H: 586.5485; found: 586.5553

S5. Synthesis and analytical data of ligand, $L_3(H)_2$



Scheme 3 Synthesis of ligand, $\mathbf{L_3}(H)_2$.

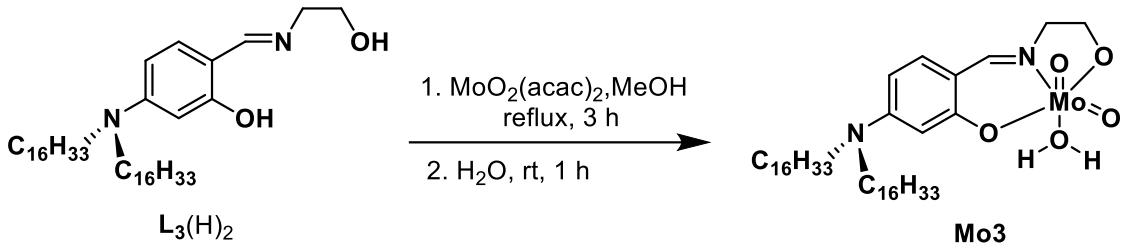
The double chain integrated Schiff base ligand $\mathbf{L_3}(H)_2$ was synthesized adopting a literature protocol with necessary modification.[4] The precursor $\mathbf{A_3}$ (1.0 g, 1.7 mmol) was taken in 15 mL methanol and ethanolamine (0.1 g, 1.7 mmol) was added to the reaction mixture followed by a few drops of acetic acid as a catalyst. The reaction mixture was stirred for 3 h under reflux conditions. After completion of the reaction the obtained clear solution was allowed to stand overnight to get a yellow crystalline solid Schiff base compound.

^{1}H NMR (400 MHz, $CDCl_3$, 24 °C): δ 8.05 (s, 1H), 6.99-6.97 (m, 1H), 6.13-6.11 (m, 1H), 6.05 (s, 1H), 3.86-3.84 (m, 2H), 3.64-3.63 (m, 2H), 3.27-3.24 (m, 4H), 1.58 (m, 4H), 1.30-126 (m, 52H), 0.89-0.86 (m, 6H) *ppm*.

$^{13}C\{^{1}H\}$ NMR (126 MHz, $CDCl_3$, 24 °C): δ 167.67, 164.48, 152.74, 133.36, 108.23, 103.59, 98.70, 62.44, 59.02, 51.19, 32.07, 29.84, 29.80, 29.75, 29.66, 29.50, 27.60, 27.25, 22.83, 14.26 *ppm*.

HRMS (ESI) m/z: calculated for $C_{41}H_{76}N_2O_2$+H: 629.5907; found: 629.5974

S6. Synthesis and analytical data of oxomolybdenum complex $MoO_2L_3(OH_2)$, Mo3



Scheme 4 Synthesis of oxomolybdenum complex $[MoO_2(\mathbf{L_3})(H_2O)]$**, Mo**3**.**

The oxomolybdenum complex $[MoO_2(\mathbf{L_3})(H_2O)]$**, Mo**3 was synthesized following a general method demonstrated in our earlier report.[1] Ligand $\mathbf{L_3}(H)_2$ (0.5 g, 0.8 mmol) and bis(acetylacetonato)dioxomolybdenum(VI) (0.26 g, 0.8 mmol) were dissolved in 15 mL methanol and stirred for 3 h under reflux condition. After completion of the reaction the obtained yellow colored solution was allowed to stand overnight. A yellow-colored semi-crystalline was observed which was filtered and dried under vacuum. The obtained yellow solid was taken in 15 mL water and stirred for 1 hour at room temperature. The precipitate was further filtered and dried under vacuum, providing the desired compound **Mo**3.

^{1}H NMR (400 MHz, $CDCl_3$, 24 ^{o}C): δ 8.31 (s, 1H), 7.16-7.14 (m, 1H), 6.31-6.29 (m, 1H), 6.19 (s, 1H), 4.63-4.60 (m, 2H), 4.16-4.13 (m, 2H), 3.31-3.28 (m, 4H), 1.31-1.26 (m, 56H), 0.89-0.86 (m, 6H) *ppm*.

^{13}C{^{1}H} NMR (126 MHz, $CDCl_3$, 24 ^{o}C): δ 164.09, 163.48, 155.45, 134.97, 109.82, 106.58, 99.88, 71.24, 63.72, 51.50, 32.07, 29.89, 29.85, 29.81, 29.74, 29.61, 29.50, 27.54, 27.20, 22.83, 14.25 *ppm*.

IR (KBr pellet) ν = 2917, 2856, 1607, 1523, 1461, 1233, 931, 897, 636 cm^{-1}.

MS (ESI) m/z: calculated for $[(\mathbf{Mo}3+H)-H_2O]^+$: 757.4786; found: 757.4768

S7. NMR spectra(^{1}H and ^{13}C) of complexes Mo1 and Mo2

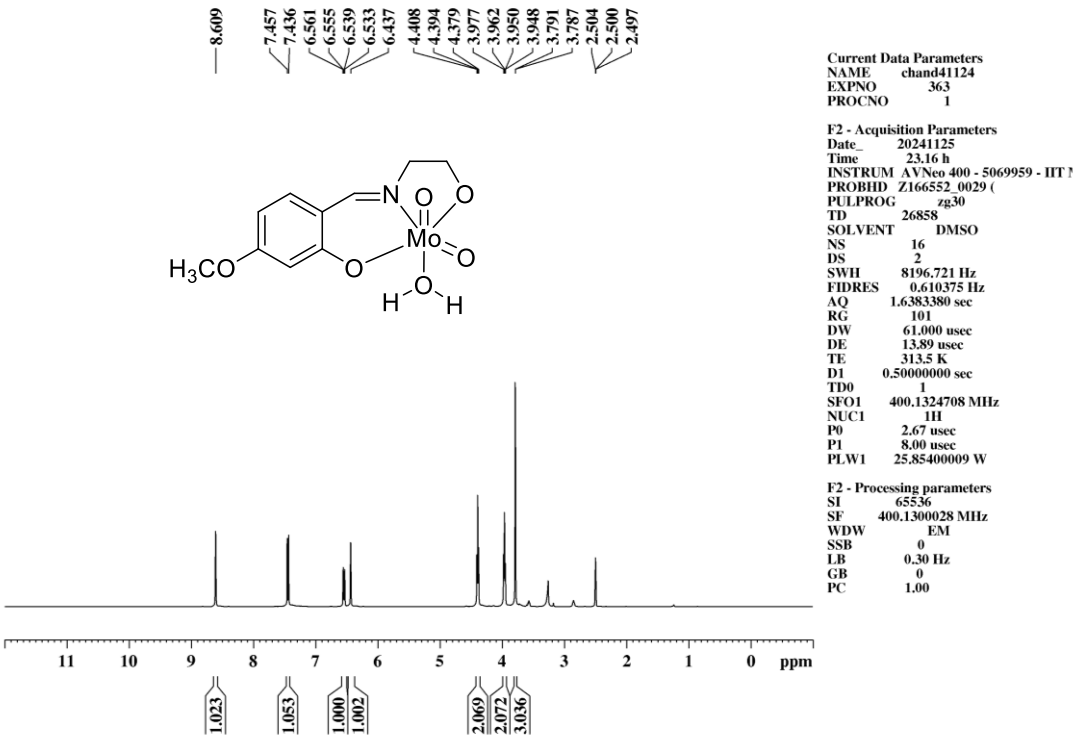


Figure S1 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of **Mo1**.

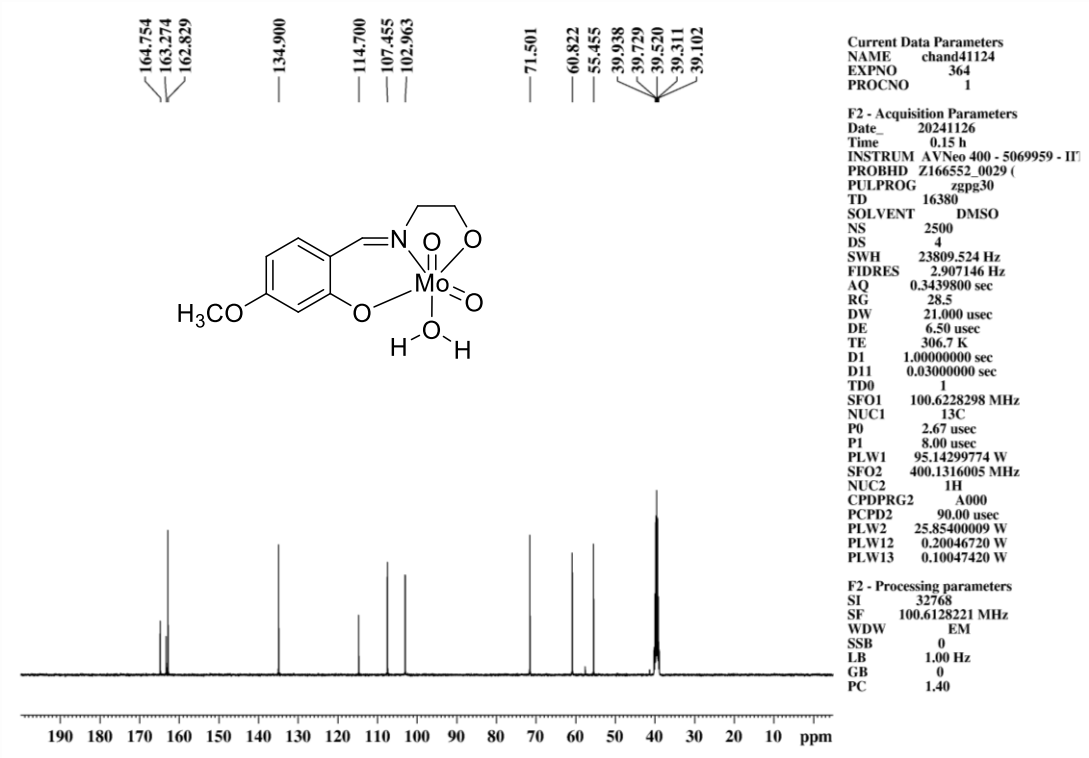


Figure S2 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of **Mo1**.

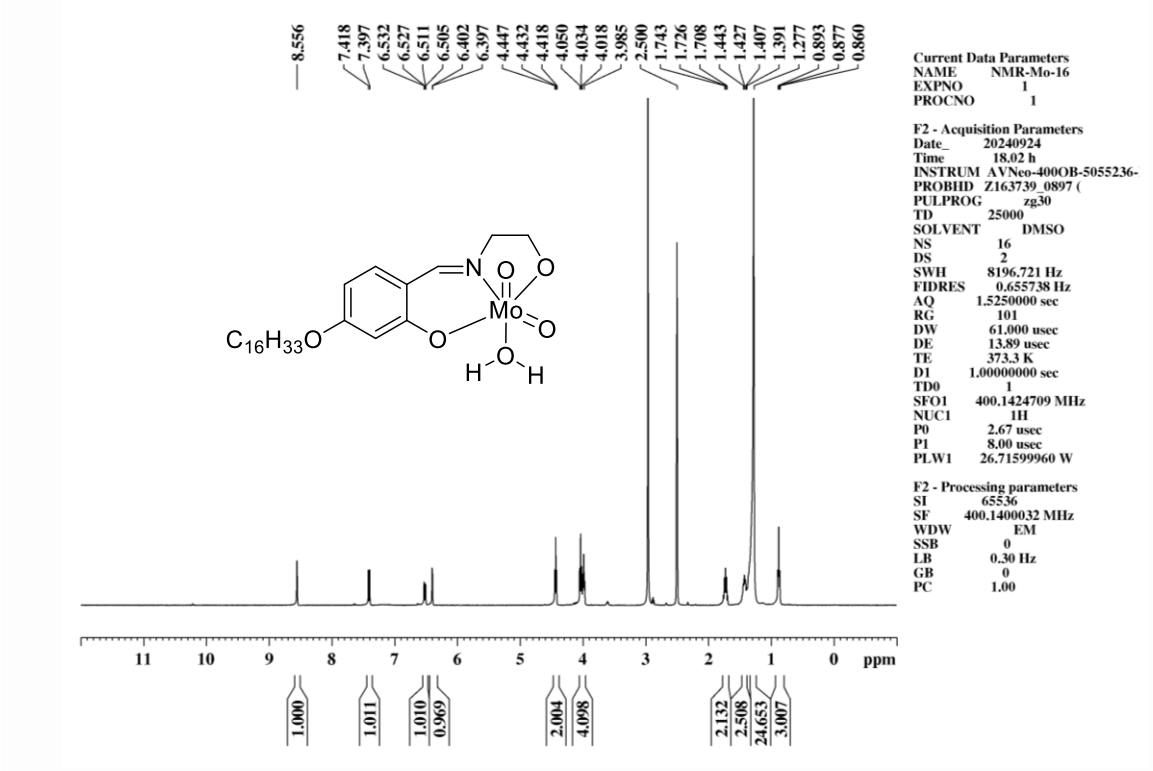


Figure S3. ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of **Mo2**.

S8. NMR spectra(^{1}H and ^{13}C) and HRMS of precursor, P_3

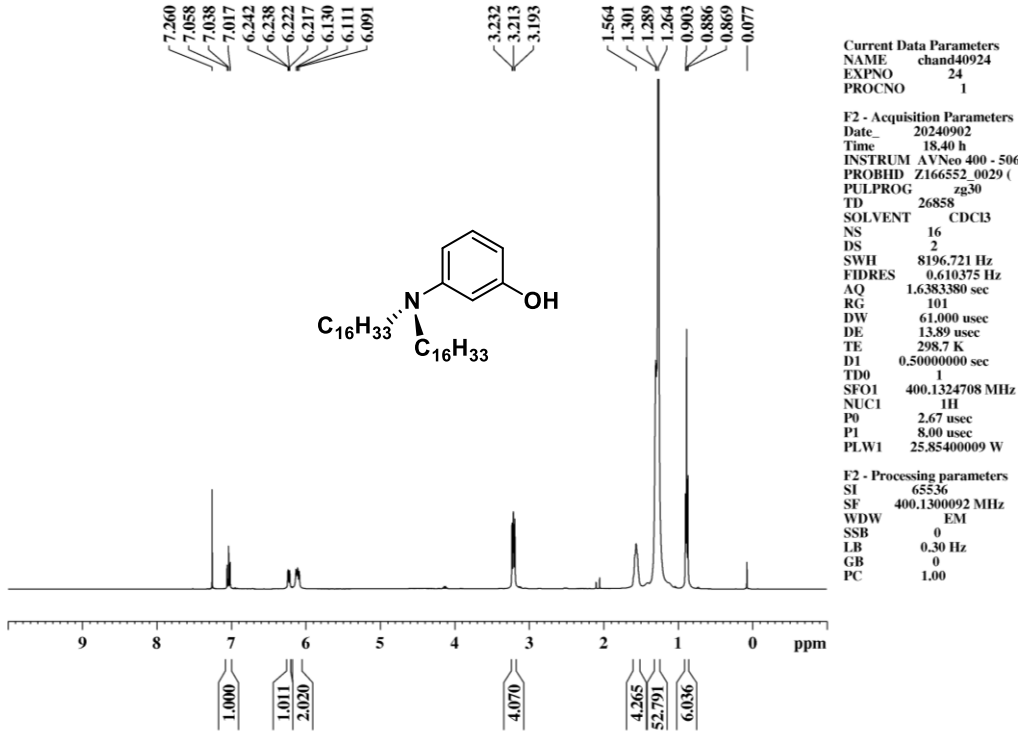


Figure S4 ^{1}H NMR spectrum (400 MHz, $CDCl_3$) of 3-(hexadecyl)aminophenol (**P_3**).

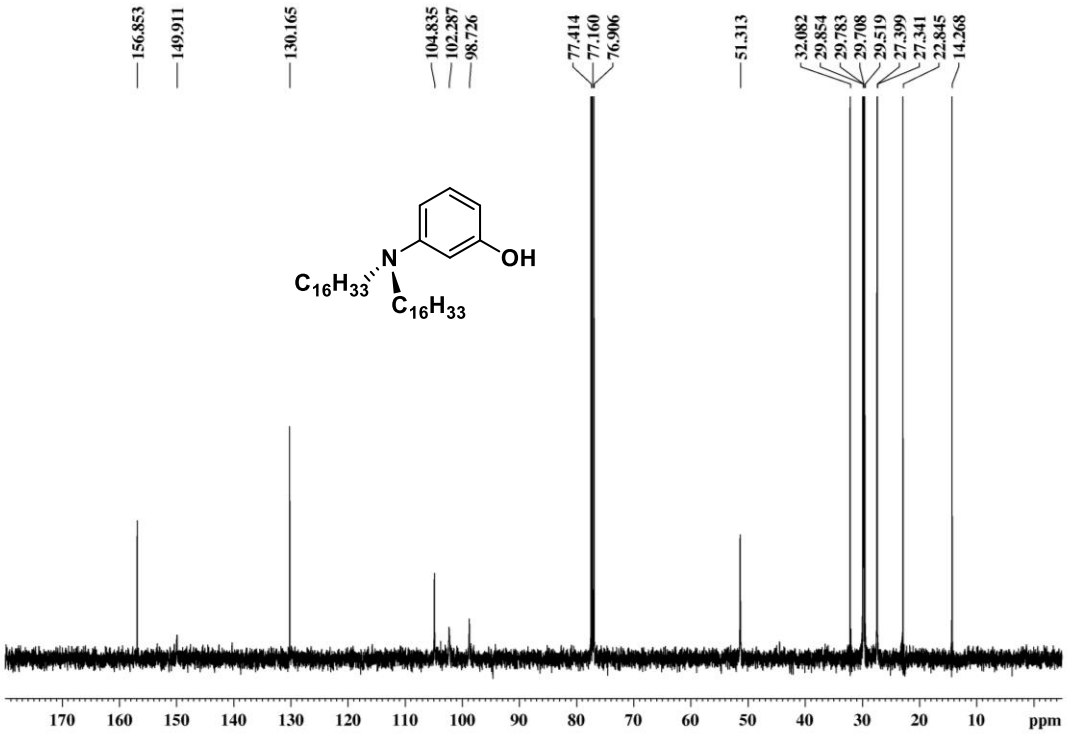


Figure S5 ^{13}C NMR spectrum (126 MHz, $CDCl_3$) of 3-(hexadecyl)aminophenol **(P_3)**.

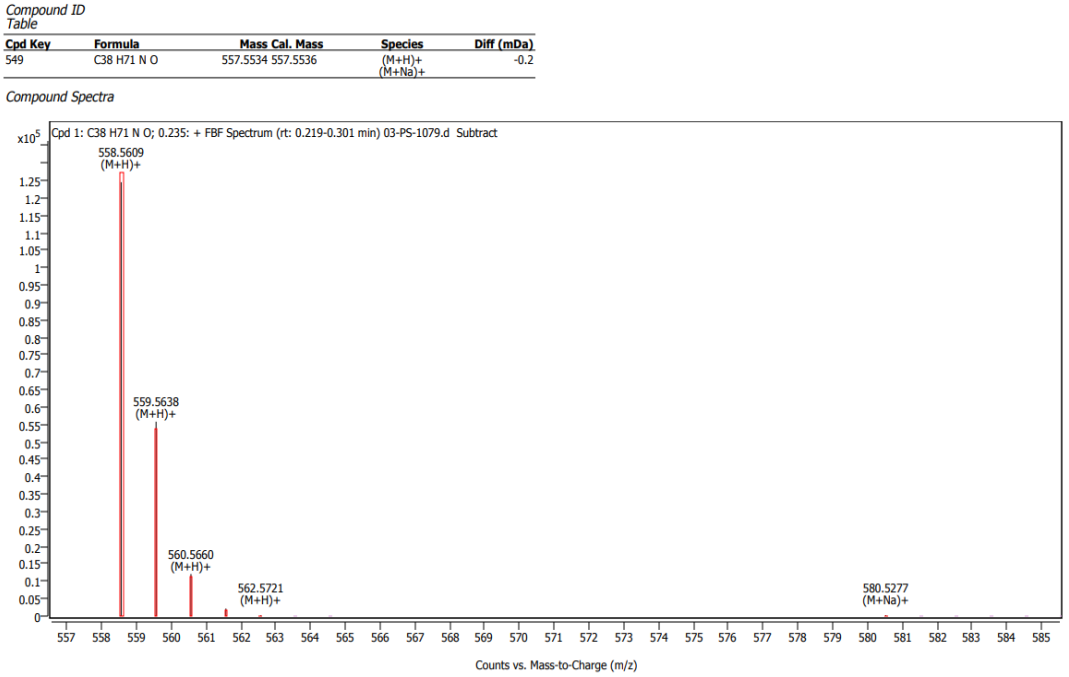


Figure S6 HRMS of 3-(hexadecyl)aminophenol **(P_3)**.

S9. NMR spectra (^{1}H and ^{13}C) and HRMS of precursor, A_3

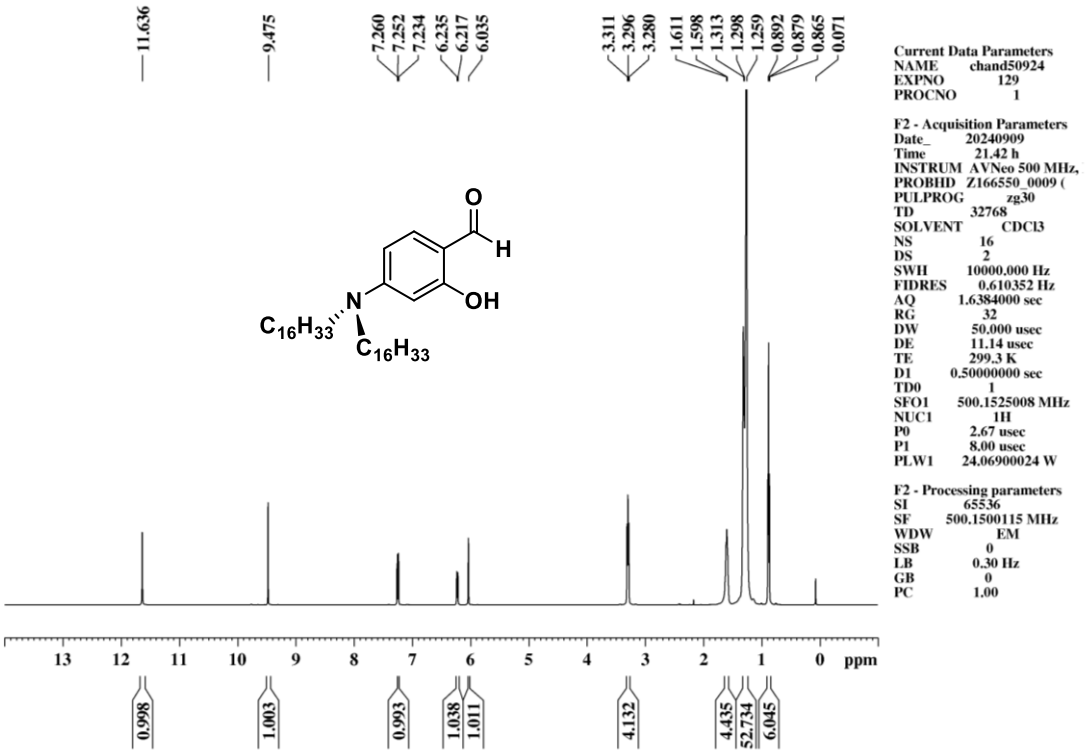


Figure S7 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of precursor **A_3**.

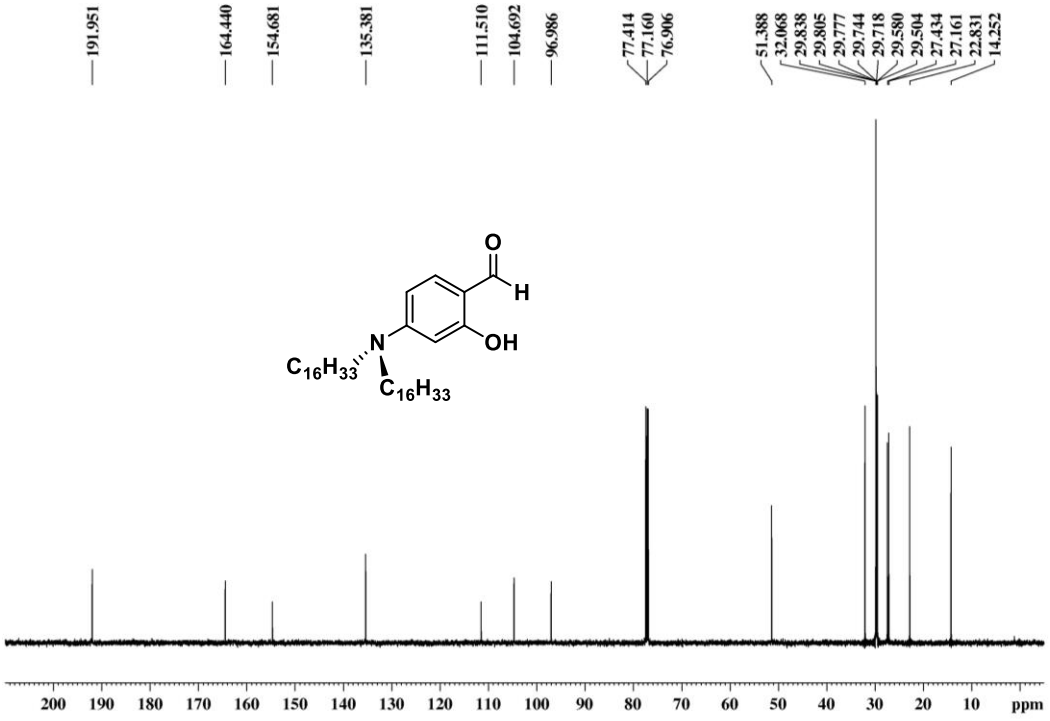


Figure S8 ^{13}C NMR spectrum (126 MHz, $CDCl_3$) of precursor **A_3**.

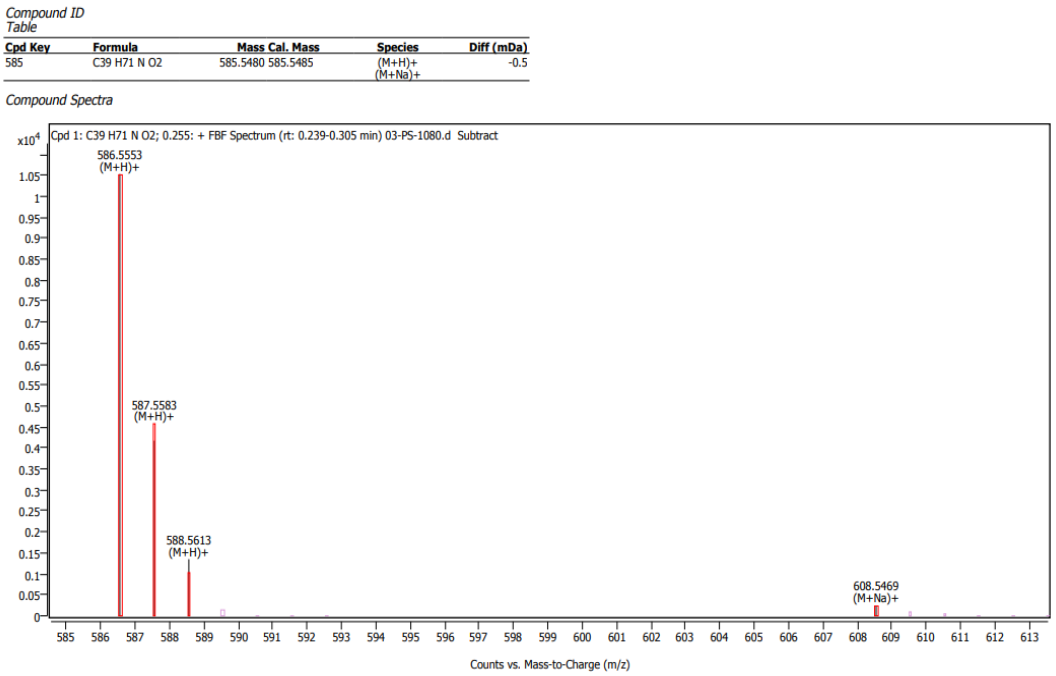


Figure S9 HRMS of precursor **A$_3$**.

S10. NMR spectra (^{1}H and ^{13}C) and HRMS of ligand, L$_3$(H)$_2$

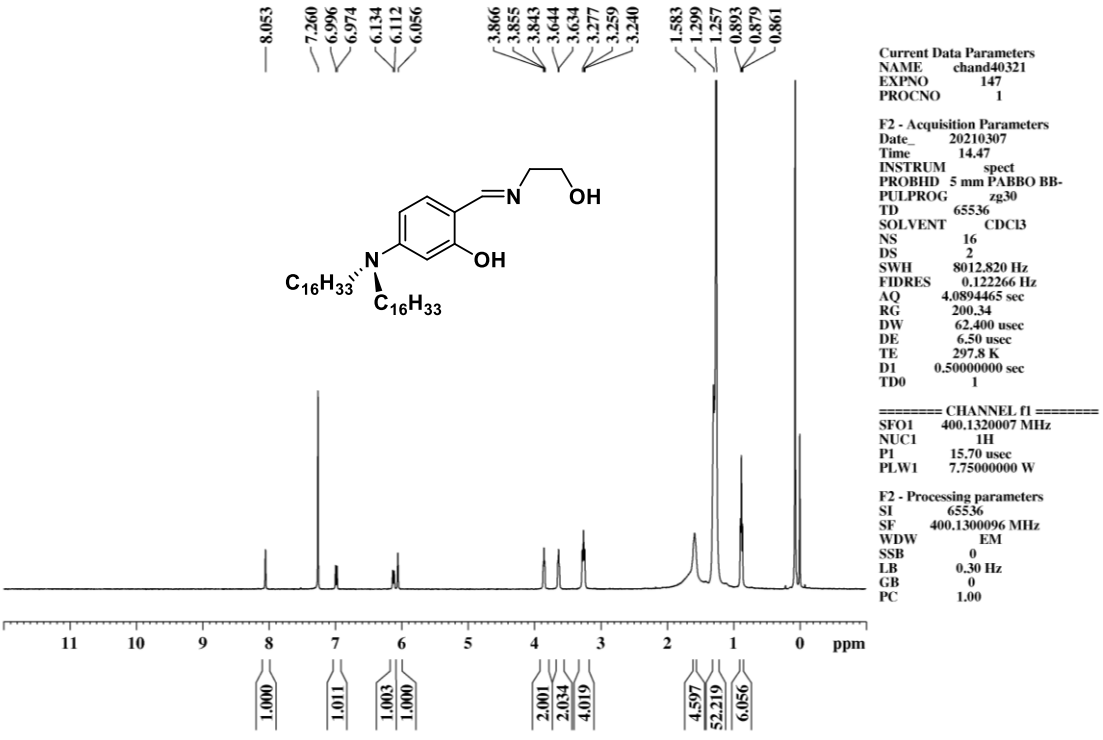


Figure S10 ^{1}H NMR spectrum (400 MHz, $CDCl_3$) of ligand **L$_3$**(H)$_2$.

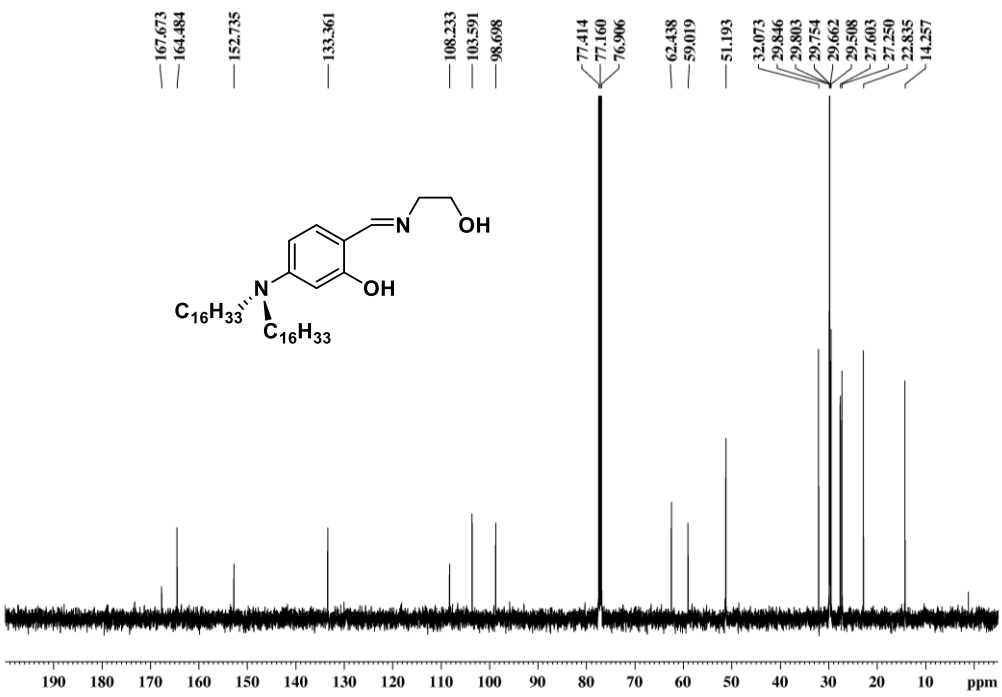


Figure S11 ^{13}C NMR spectrum (126 MHz, $CDCl_3$) of ligand $\mathbf{L_3}(H)_2$.

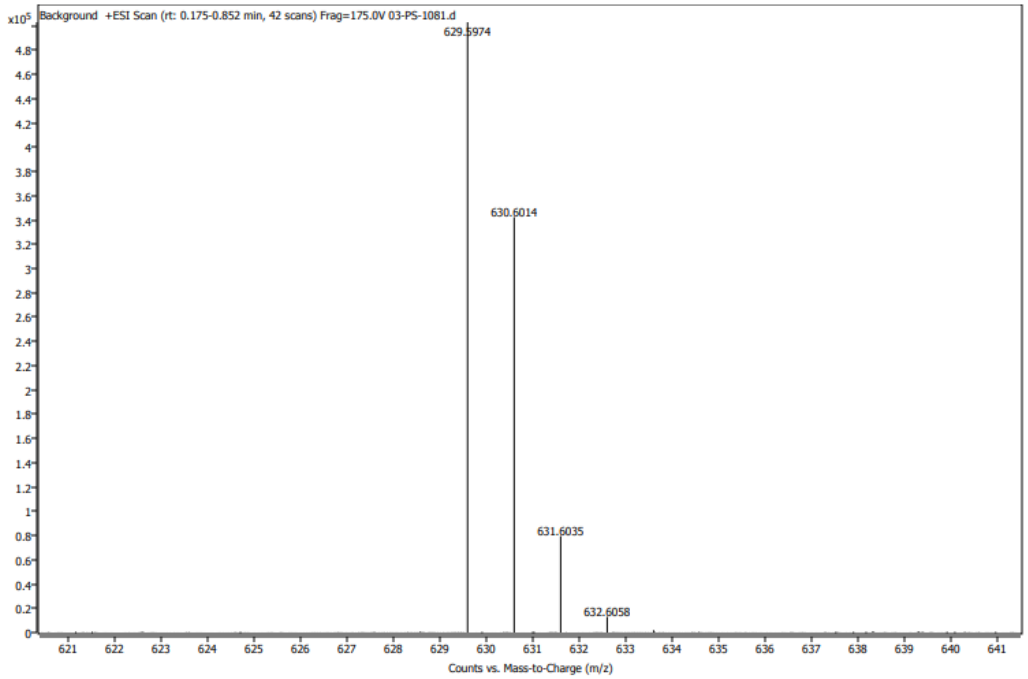


Figure S12 HRMS of ligand $\mathbf{L_3}(H)_2$.

S11. NMR spectra (^{1}H and ^{13}C), ESI-MS, FTIR spectrum and energy minimized structure of the oxomolybdenum complex Mo3

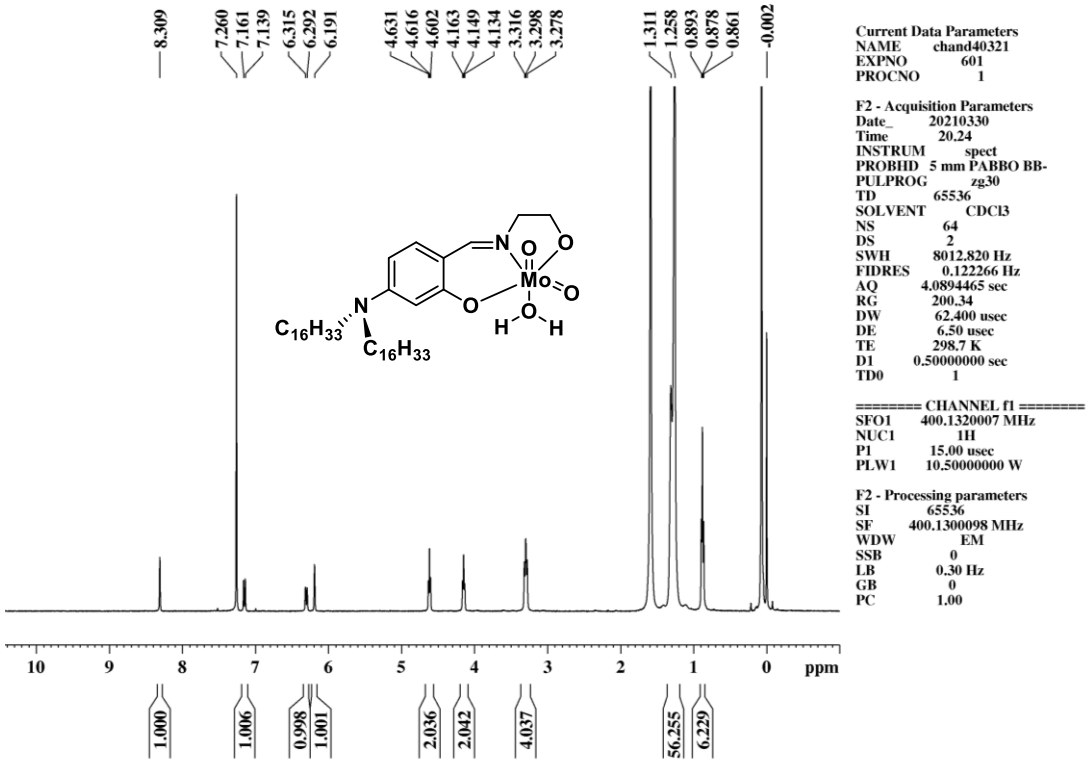


Figure S13 ^{1}H NMR spectrum (400 MHz, 25 °C, $CDCl_3$) of $[MoO_2(\mathbf{L_3})(H_2O)]$, **Mo3**.

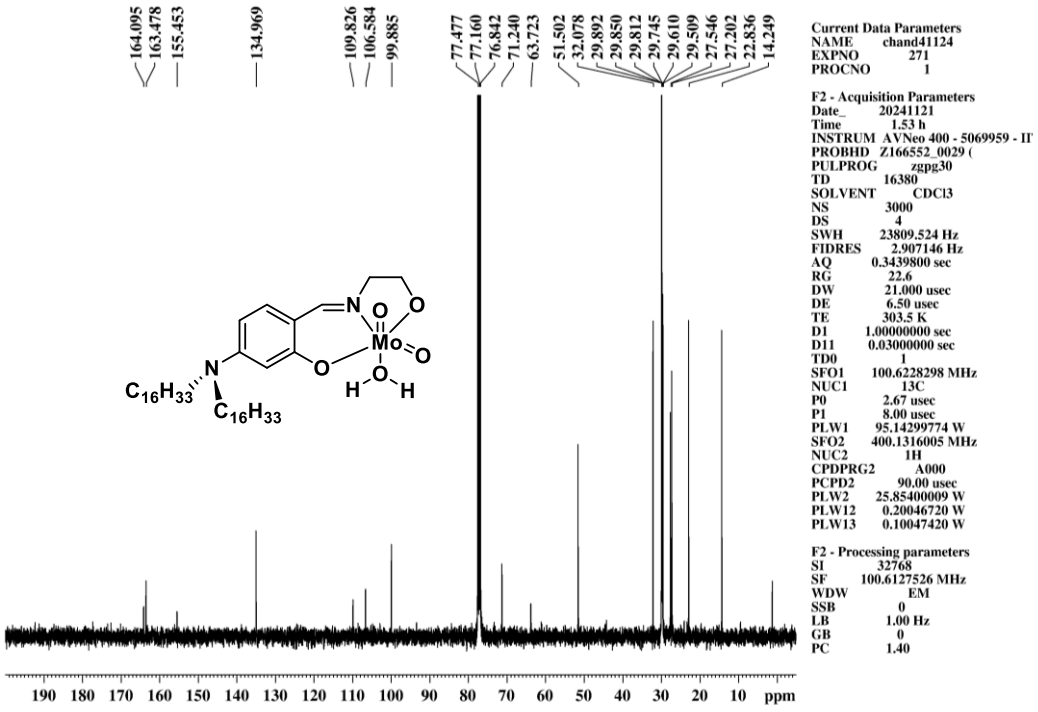


Figure S14 ^{13}C NMR spectrum (101 MHz, 25 °C, $CDCl_3$) of $[MoO_2(\mathbf{L_3})(H_2O)]$, **Mo3**.

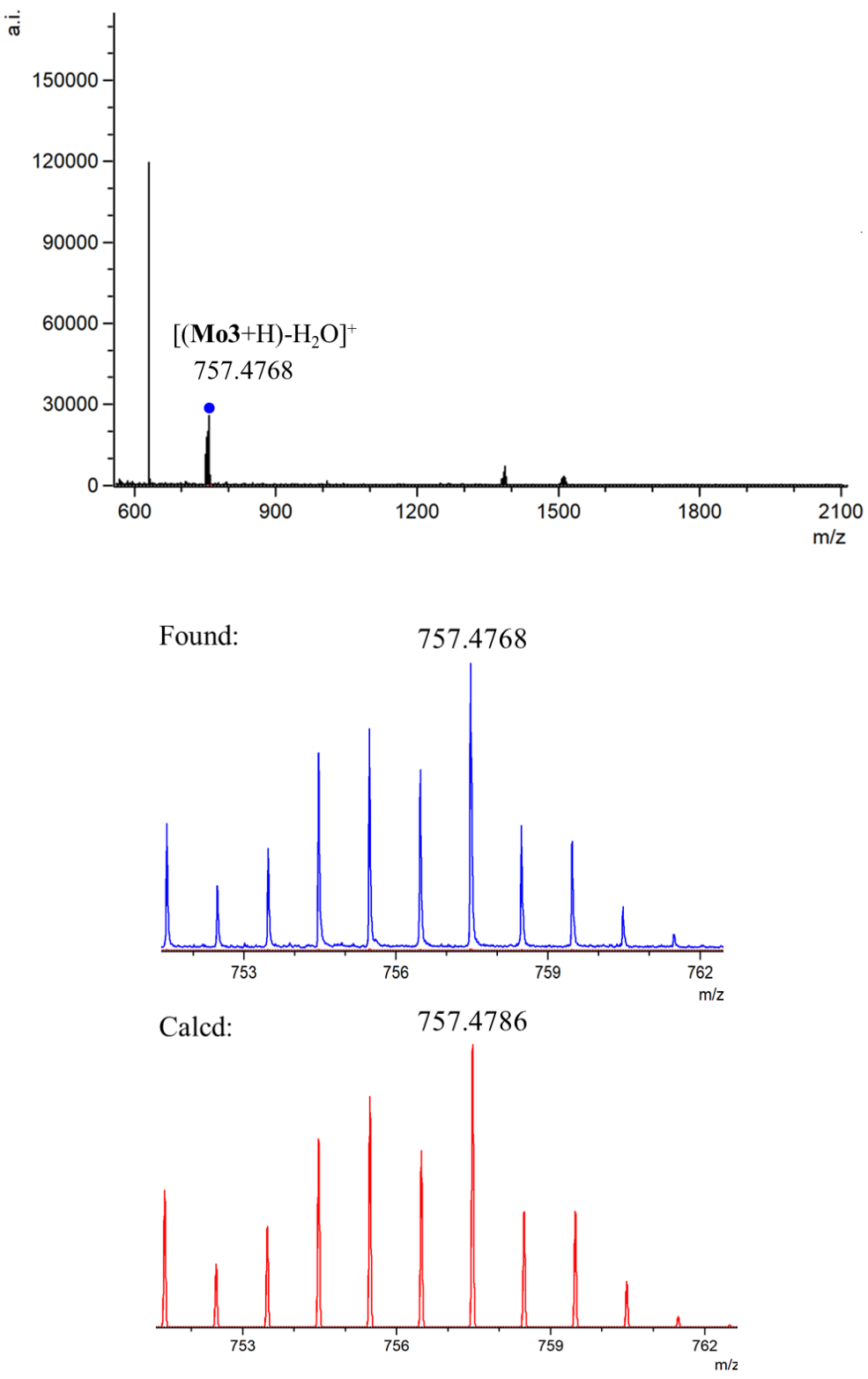


Figure S15 ESI-MS of complex **Mo3**.

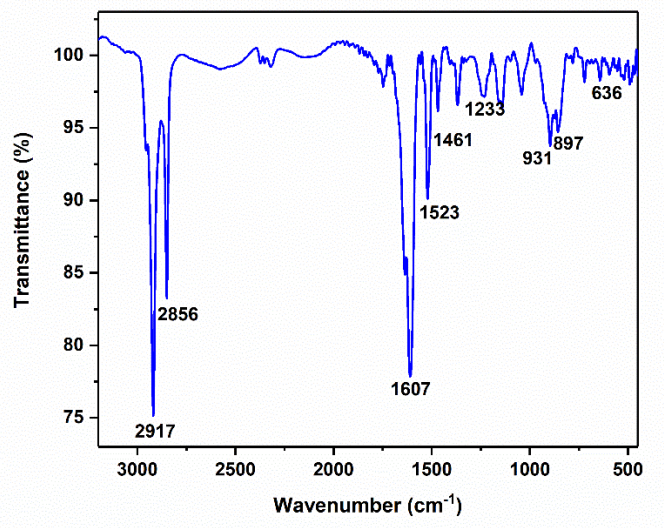


Figure S16 FT-IR spectrum of complex **Mo3**.

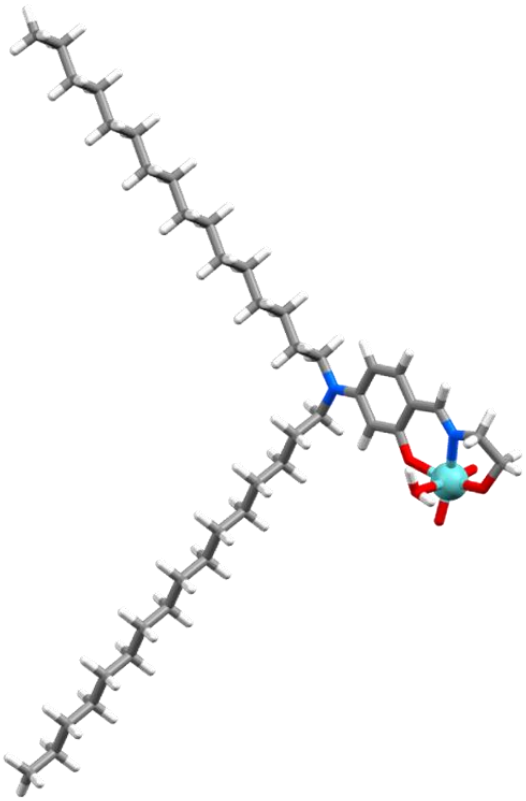


Figure S17 Energy minimized structure of [MoO_2(**L_3**)(H_2O)], **Mo3** (in gas phase).

S12. Conductivity study of oxomolybdenum complexes, Mo2 and Mo3

Electrical conductivity method was employed to determine the critical micelle concentration (CMC) of the complexes **Mo2** and **Mo3**. The conductivity experiments were carried out in a double-jacket flask. The temperature of the flask was maintained at 25 °C by circulating water with Julabo FP50 Refrigerated - Heating Circulators. LMCM-20 conductivity meter from LABMAN scientific instruments was used for the determination of critical micellar concentration of the complexes. Solutions were prepared in deionized water which was first filtered with a Millipore Milli-Q system. A step-by-step dilution-extraction method was adopted for the measurements of specific conductance of the complex at various concentrations in order to avoid dilution error.[5,6] The conductance was plotted as a function of molar concentration and the observed values of CMC of the complexes **Mo2** and **Mo3** are given in Figure S18.

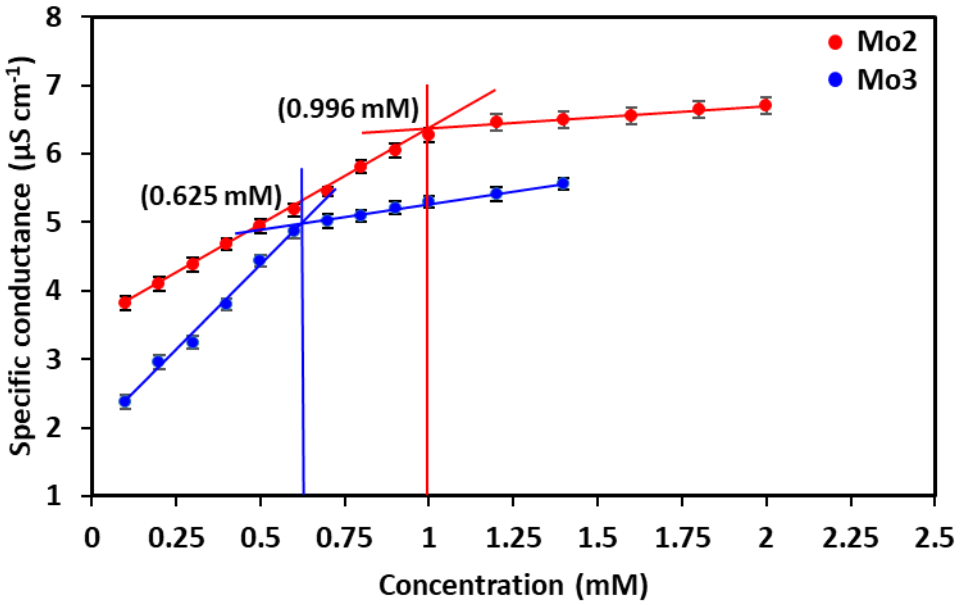


Figure S18 Conductivity study of oxomolybdenum complexes, **Mo2** and **Mo3** to determine critical micelle concentration (CMC).

S13. General procedure for the oxidative coupling of benzylamines with 1,2-diaminobenzenes (For isolation and kinetics study)

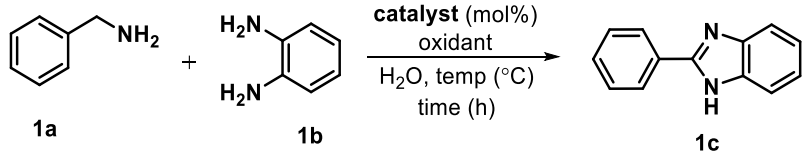
A mixture of benzylamine (0.9 mmol), 1,2-diaminobenzene (0.75 mmol), and catalyst (1.5 mol%) was taken in 0.5 mL water in a ~12.0 mL reaction tube (~5.0 × 2.0 cm of height × width) fitted with a reflux condenser. The reaction mixture was stirred for 18 h at 80 ºC temperature under open air atmosphere. The progress of the reaction was monitored by TLC. After completion of the reaction, 0.5 mL water was added to the reaction mixture, and the aqueous layer was extracted with ethyl acetate (3 × 1 mL). The organic layer containing benzimidazole was dried over anhydrous sodium sulfate and evaporated under reduced pressure. The desired product was purified by column chromatography (silica gel) using hexane-ethyl acetate as an eluent. The purified product was characterized by ^{1}H and ^{13}C NMR spectroscopy.

For kinetics study: The catalyzed reactions were allowed to proceed up to a pre-decided time and then quenched by adding ethyl acetate. The samples containing crude solid were obtained after evaporation and directly used to record ^{1}H NMR to calculate the percentage of conversion. (Fig. 7 of Manuscript and Fig S19 and S20 of Supp Info).

For substrate scope: The above-mentioned general method was followed to study substrate scope using catalyst **Mo3**.

S14. Optimization of reaction conditions for oxidative coupling of benzylamine with 1,2-diaminobenzene

Table S1 Optimization of reaction condition for the oxidative coupling of benzylamine, **1a** with 1,2-diaminobenzene, **1b** in water.[a]



1a 1b 1c

Entry	Catalysts (x mol%)	Oxidant	Temp (°C)	Time	Yield (%)[b]
1	-	open air	rt	24	-
2	-	open air	50	24	<05
3	**Mo1** (1.0 mol%)	open air	50	24	11
4	**Mo2** (1.0 mol%)	open air	50	24	35
5	**Mo3** (1.0 mol%)	open air	50	24	46

6	**Mo3** (1.0 mol%)	open air	80	24	83
7	**Mo3** (1.0 mol%)	open air	100	24	87
8	**Mo3** (1.5 mol%)	open air	80	24	94
9	**Mo3 (1.5 mol%)**	**open air**	**80**	**18**	**92**
10	**Mo3** (1.5 mol%)	open air	80	12	79
11	**Mo2 (1.5 mol%)**[c]	**open air**	**80**	**18**	**73**
12	**Mo1** (1.5 mol%)	open air	80	18	28
13	-	open air	80	18	11
14	Only $\mathbf{L_3}(H)_2$	open air	80	18	14

[a]Reaction conditions: benzylamine, **1a** (0.9 mmol) and 1,2-diaminobenzene, **1b** (0.75 mmol) in 0.5 mL water under oxygen atmosphere at a given temperature (oil bath temperature). [b]Isolated yield. [c]**1c** was isolated in 84% upon increasing the catalyst loading to 2.5 mol%.

The reaction conditions for the targeted organic reaction were optimized taking benzylamine, **1a** and 1,2-diaminobenzene, **1b** as model substrates, varying the catalysts, catalyst loading, oxidant, temperature, and time. The acquired results are summarized in Table S1.

We began our investigation by treating benzylamine, **1a** (0.9 mmol) with 1,2-diaminobenzene, **1b** (0.75 mmol) in 0.5 mL water, in the absence of catalyst, at room temperature for 24 hours, under open air conditions. No product formation was observed (Table S1, entry 1). However, when the reaction temperature was raised to 50 °C, a small amount of 2-phenyl-1*H*-benzimidazole, **1c** (<05 % yield) was observed (Table S1, entry 2). Based on this observation, we introduced 1.0 mol% of non-micellar catalyst, **Mo1** to the reaction mixture, and heated it at 50 °C for 24 hours, resulting in a slight increment of product yield to 11% (Table S1, entry 3). Notably, the micellar catalysts with a single chain, **Mo2** (1.0 mol%), and double chain, **Mo3** (1.0 mol%) in ligand backbone were employed under similar conditions, the yield of **1c** increased significantly to 35% and 46%, respectively (Table S1, entries 4 and 5). These results underscore the crucial role of the surfactant entity within the metal complex, which forms a metallomicelle during the reaction process, facilitating the reaction in a water medium. Proven the superior reactivity of **Mo3**, the double chain-based catalyst, **Mo3**, over **Mo2** and non-micellar catalyst, **Mo1**, further optimization was conducted using **Mo3**.

To achieve a better yield of the desired product, the reaction temperature was increased to 80 °C, and **1c** was isolated in 83% yield (Table S1, entry 6). A further increase in the reaction temperature to 100 °C did not resulted a significant change in the product yield, with **1c** being

isolated at 87% (Table S1, entry 7). Notably, when the catalyst loading was increased to 1.5 mol% and the reaction was conducted at 80 °C for 24 hours, complete conversion was achieved, and product **1c** was isolated in 94% yield (Table S1, entry 8). Reducing the reaction time to 18 and 12 hours led to isolated yields of 92 and 79%, respectively (Table S1, entries 9 and 10). Thus, entry 9 in Table S1 was considered as the optimized reaction condition for the oxidative coupling of benzylamine, **1a** with 1,2-diaminobenzene, **1b** to form 2-substituted benzimidazole, **1c** catalyzed by **Mo3**. Under these reactions, the catalytic ability of **Mo2** was tested, resulting in only 73% of benzimidazole **1c** was obtained (Table S1, entry 11). When the catalyst loading of **Mo2** was increased to 2.5 mol%, **1c** was isolated in 84% yield. Only 28% and 11% yield of benzimidazole **1c** was isolated when the reactions carried out using **Mo1** and in absence of catalyst under optimized reaction conditions (Table S1, entries 12 and 13). In presence of ligand $\mathbf{L_3}(H)_2$ the yield **1c** was 14% (Table S1, entry 14).

S15. Reaction kinetics of benzimidazole formation by Mo2 and Mo3 catalyzed oxidative coupling reactions

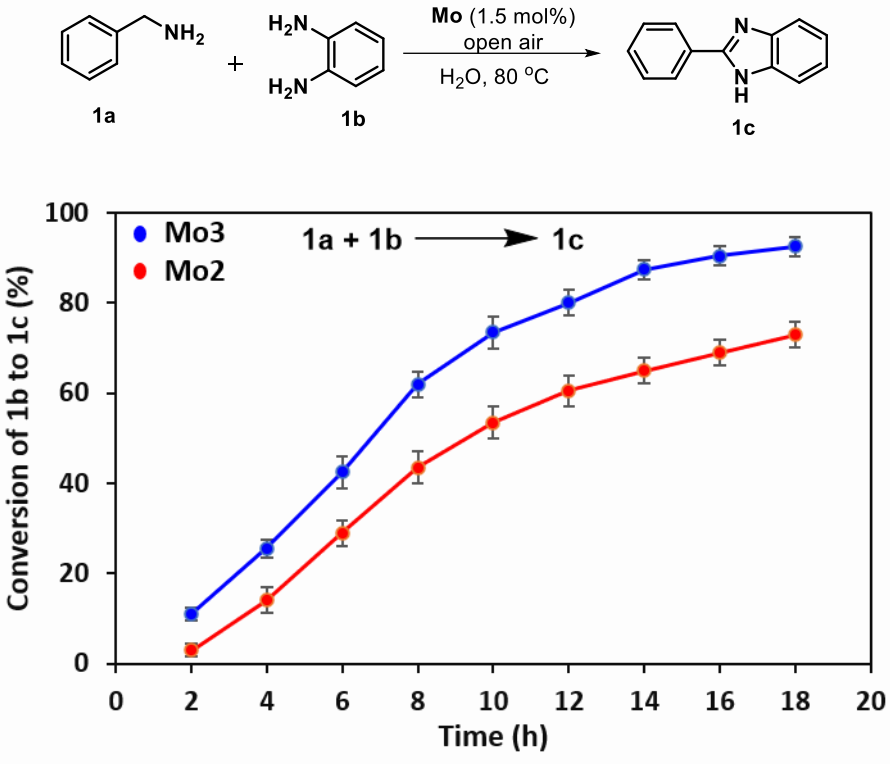


Figure S19. Conversion plot of 1,2-diaminobenzene, **1b** to corresponding benzimidazole, **1c** at 2 h time intervals catalyzed by **Mo2** and **Mo3**.

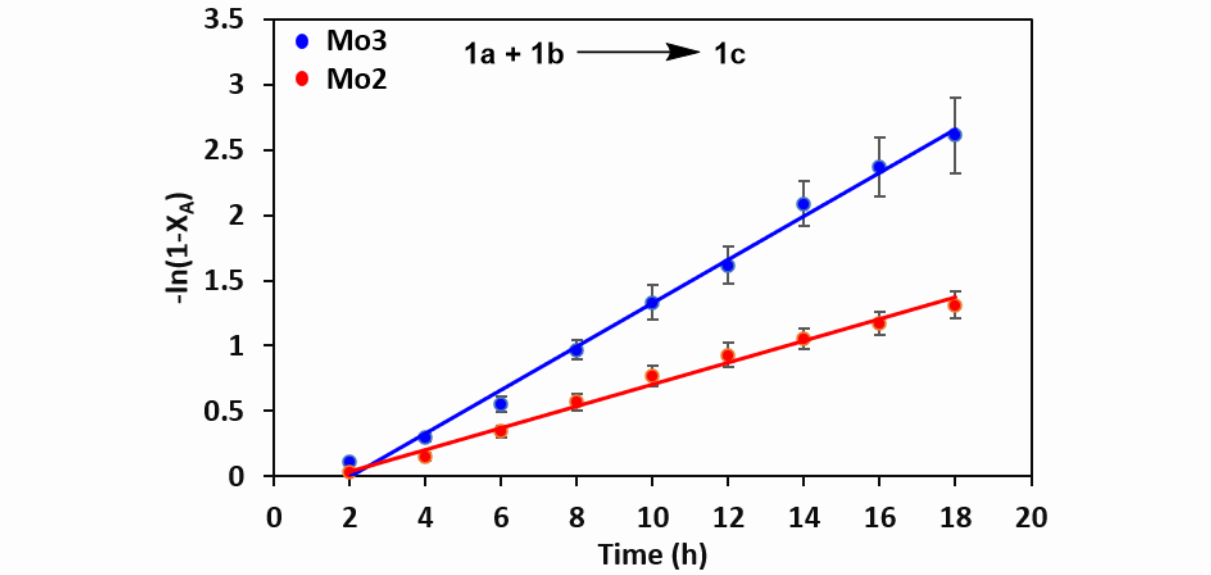
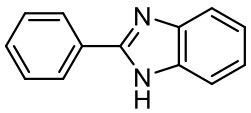


Figure S20. Plot of -ln(1-X_A) vs. time for the oxidative coupling of benzylamine, **1a** and 1,2-diaminobenzene, **1b** to corresponding benzimidazole, **1c** at 2 h time intervals catalyzed by **Mo2** and **Mo3**. The slope values (*k'*) for **Mo2 and Mo3** are obtained from the graph and found to be 0.083 and 0.166 (h^{-1}), and corresponding second-order rate constants are calculated as 3.68 and 7.38 $M^{-1}h^{-1}$ respectively.

S16. Analytical data of the benzimidazoles

(1) 2-phenyl-1*H*-benzimidazole (**1c**):

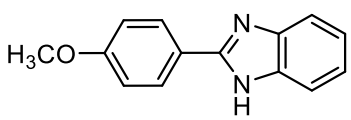


The title compound **1c** was obtained following the general method for oxidative coupling using commercially available benzylamine, **1a** and 1,2-diaminobenzene, **1b** affording the title compound as yellow solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.88 (bs, 1H), 8.19-8.17 (m, 2H), 7.57-7.49 (m, 5H), 7.21-7.20 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 151.18, 130.15, 129.76, 128.87, 126.45, 126.39, 121.69, 118.78, 111.32 *ppm.*

(2) 2-(4-methoxyphenyl)-1*H*-benzimidazole (**2c**):



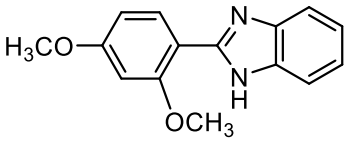
The title compound **2c** was obtained following the general method for oxidative coupling using commercially available 4-methoxybenzylamine, **2a** and 1,2-diaminobenzene, **1b**

affording the title compound as white solid, compound data matches those previously reported.[7]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 12.72 (bs, 1H), 8.12-8.10 (m, 2H), 7.55 (bs, 2H), 7.17-7.16 (m, 2H), 7.12-7.10 (d, 2H, J = 8.0 Hz), 3.84 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 160.59, 151.32, 127.98, 122.69, 121.79, 114.35, 55.32 *ppm*.

(3) 2-(2,4-dimethoxyphenyl)-1*H*-benzimidazole (**3c**):

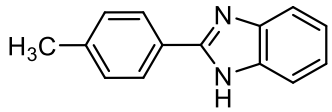


The title compound **3c** was obtained following the general method for oxidative coupling using commercially available 2,4-dimethoxybenzylamine, **3a** and 1,2-diaminobenzene, **1b** affording the title compound as light yellow solid, compound data matches those previously reported.[8]

^{1}H NMR (400 MHz, $CDCl_3$, 24 °C): δ 8.50-8.48 (m, 1H), 7.60 (bs, 2H), 7.23-7.21 (m, 2H), 6.66-6.64 (m, 1H), 6.54 (s, 1H), 3.98 (s, 3H), 3.84 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, $CDCl_3$, 24 °C): δ 162.43, 158.18, 150.21, 131.41, 122.30, 111.04, 106.05, 98.92, 55.98, 55.62 *ppm*.

(4) 2-(p-tolyl)-1*H*-benzimidazole (**4c**):

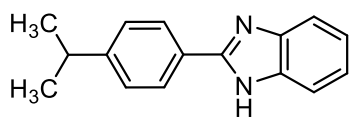


The title compound **4c** was obtained following the general method for oxidative coupling using commercially available 4-methylbenzylamine, **4a** and 1,2-diaminobenzene, **1b** affording the title compound as yellow solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.84 (bs, 1H), 8.09-8.07 (m, 2H), 7.62-7.55 (m, 2H), 7.36-7.34 (m, 2H), 7.20-7.18 (m, 2H), 2.37 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 151.42, 139.57, 129.53, 127.48, 126.41, 122.29, 121.64, 118.74, 111.29, 20.98 *ppm*.

(5) 2-(4-isopropylphenyl)-1*H*-benzimidazole (**5c**):

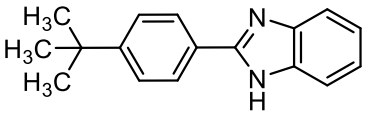


The title compound **5c** was obtained following the general method for oxidative coupling using commercially available 4-isopropylbenzylamine, **5a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[9]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.76 (bs, 1H), 8.11-8.09 (m, 2H), 7.58 (bs, 2H), 7.43-7.41 (m, 2H), 7.20-7.17 (m, 2H), 3.00-2.93 (m, 1H), 1.26 (s, 3H), 1.24 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 151.29, 150.24, 129.36, 127.79, 126.76, 126.43, 121.69, 33.25, 23.57 *ppm*.

(6) 2-(4-*tert*-butylphenyl)-1*H*-benzimidazole (**6c**):

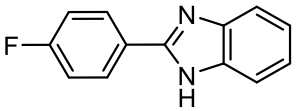


The title compound **6c** was obtained following the general method for oxidative coupling using commercially available 4-*t*-butylbenzylamine, **6a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.79 (bs, 1H), 8.11-8.09 (m, 2H), 7.66-7.64 (m, 1H), 7.58-7.55 (m, 2H), 7.52-7.50 (m, 1H), 7.20-7.17 (m, 2H), 1.33 (s, 9H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 152.48, 151.22, 143.81, 134.91, 127.41, 126.17, 125.62, 122.22, 121.43, 118.66, 111.10, 34.51, 30.91 *ppm*.

(7) 2-(4-fluorophenyl)-1*H*-benzimidazole (**7c**):

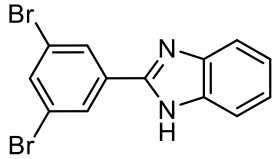


The title compound **7c** was obtained following the general method for oxidative coupling using commercially available 4-fluorobenzylamine, **7a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.86 (bs, 1H), 8.24-8.20 (m, 2H), 7.61 (bs, 2H), 7.41-7.37 (m, 2H), 7.21 (s, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 164.22, 161.76, 150.35, 150.32, 128.69, 128.61, 126.78, 122.02, 118.80, 115.97, 115.76, 111.21 *ppm*.

(8) 2-(3,5-Dibromophenyl)-1*H*-benzimidazole (**8c**):

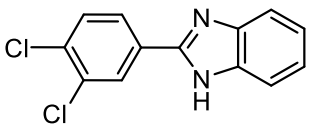


The title compound **8c** was obtained following the general method for oxidative coupling using 3,5-dibromobenzylamine, **8a** and 1,2-diaminobenzene, **1b** affording the compound as light brown solid.

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 13.05 (bs, 1H), 8.36-8.35 (m, 2H), 7.92-7.91 (m, 1H), 7.68-7.56 (m, 2H), 7.24 (s, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 148.09, 134.10, 133.80, 127.95, 123.22, 122.98, 122.09, 119.17, 111.58 *ppm*.

(9) 2-(3,4-dichlorophenyl)-1H-benzoimidazole (**9c**):

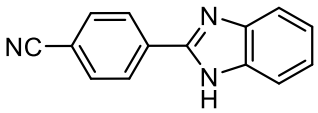


The title compound **9c** was obtained following the general method for oxidative coupling using 3,4-dichlorobenzylamine, **9a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid.[10]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 13.03 (bs, 1H), 8.39-8.39 (m, 1H), 8.16-8.13 (m, 1H), 7.82-7.80 (m, 1H), 7.69-7.67 (m, 1H), 7.56-7.54 (m, 1H), 7.23 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 148.80, 143.58, 134.95, 132.19, 131.76, 130.67, 127.90, 126.34, 123.01, 121.95, 119.05, 111.46 *ppm*.

(10) 4-(1*H*-benzo[*d*]imidazol-2-yl)benzonitrile (**10c**):

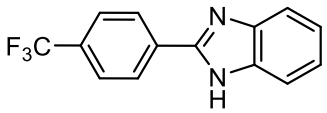


The title compound **8c** was obtained following the general method for oxidative coupling using prepared 4-cyanobenzylamine, **10a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 13.11 (bs, 1H), 8.34 (d, 2H, J = 8.5 Hz), 7.99 (d, 2H, J = 8.5 Hz), 7.70-7.58 (m, 2H), 7.25 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 149.29, 143.74, 135.07, 134.20, 132.77, 126.89, 123.25, 122.08, 119.26, 118.45, 111.80, 111.59 *ppm*.

(11) 2-(4-(trifluoromethyl)phenyl)-1*H*-benzimidazole (**11c**):

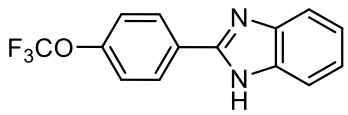


The title compound **11c** was obtained following the general method for oxidative coupling using 4-trifluoromethylbenzylamine, **11a** and 1,2-diaminobenzene, **1b** affording the title compound as yellow solid.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 13.09 (bs, 1H), 8.39-8.37 (m, 2H), 7.93-7.91 (m, 2H), 7.72-7.56 (m, 2H), 7.27-7.23 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 149.53, 134.99, 133.88, 129.38, 126.94, 125.75, 123.03, 121.91, 119.13, 111.49 *ppm*.

(12) 2-(4-(trifluoromethoxy)phenyl)-1*H*-benzimidazole (**12c**):

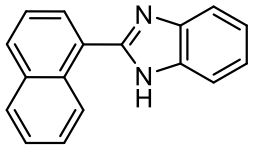


The title compound **12c** was obtained following the general method for oxidative coupling using 4-trifluoromethoxybenzylamine, **12a** and 1,2-diaminobenzene, **1b** affording the title compound as yellow solid.[11]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.95 (bs, 1H), 8.31-8.28 (m, 2H), 7.56-7.54 (m, 4H), 7.23-7.22 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 149.83, 149.17, 129.49, 129.36, 128.99, 128.33, 121.27, 120.76, 120.48, 118.70, 111.39 *ppm*.

(13) 2-(naphthalen-1-yl)-1*H*-benzimidazole (**13c**):

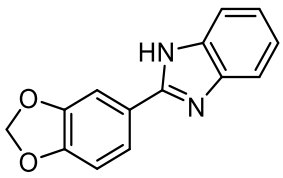


The title compound **13c** was obtained following the general method for oxidative coupling using naphthalen-1-ylmethanamine, **13a** and 1,2-diaminobenzene, **1b** affording the title compound as a white solid.[12]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 12.92 (bs, 1H), 9.12-9.10 (m, 1H), 8.10-8.09 (m, 1H), 8.05-8.01 (m, 2H), 7.79-7.77 (m, 1H), 7.70-7.61 (m, 4H), 7.28-7.26 (m, 2H) *ppm.*

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 151.34, 143.88, 134.42, 133.60, 130.49, 130.13, 128.37, 127.84, 127.51, 127.04, 126.33, 125.26, 122.62, 121.58, 119.06, 111. 33 *ppm*.

(14) 2-(benzo[*d*][1,3]dioxol-5-yl)-1*H*-benzimidazole (**14c**):

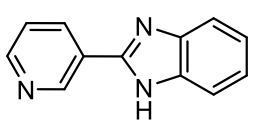


The title compound **14c** was obtained following the general method for oxidative coupling using commercially available piperonylamine, **14a** and 1,2-diaminobenzene, **1b** affording the title compound as yellow solid, compound data matches those previously reported.[10]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.68 (bs, 1H), 7.74-7.69 (m, 2H), 7.62-7.50 (m, 2H), 7.18-7.17 (m, 2H), 7.10-7.08 (m, 1H), 6.12 (s, 2H) *ppm*.

^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 151.07, 148.63, 147.78, 143.70, 134.86, 124.25, 122.12, 121.44, 120.81, 118.50, 110.96, 108.61, 106.43, 101.47 *ppm*.

(15) 2-(pyridin-2-yl)-1*H*-benzimidazole **(15c)**:

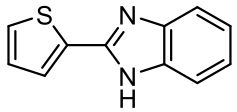


The title compound **15c** was obtained following the general method for oxidative coupling using commercially available pyridin-3-

ylmethanamine, **15a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[12]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 13.11 (bs, 1H), 9.36-9.35 (m, 1H), 8.68-8.67 (m, 1H), 8.51-8.49 (m, 1H), 7.69 (bs, 1H), 7.59-7.57(m, 2H), 7.24 (s, 2H) *ppm.*
^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 150.50, 148.86, 147.52, 143.71, 134.94, 126.16, 124.01, 122.98, 121.98, 119.09, 111.53 *ppm*.

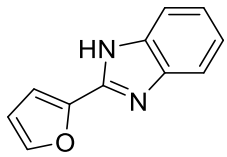
(16) 2-(thiophen-2-yl)-1*H*-benzimidazole **(16c)**:



The title compound **16c** was obtained following the general method for oxidative coupling using commercially available 2-thiophenemethylamine, **16a** and 1,2-diaminobenzene, **1b** affording the title compound as white solid, compound data matches those previously reported.[7]

^{1}H NMR (400 MHz, DMSO-d_6, 24 °C): δ 12.85 (bs, 1H), 7.83-7.82 (m, 1H), 7.71-7.70 (m, 1H), 7.54 (bs, 2H), 7.24-7.18 (m, 3H) *ppm.*
^{13}C {^{1}H} NMR (101 MHz, DMSO-d_6, 24 °C): δ 146.90, 133.61, 132.99, 128.51, 128.06, 126.52, 122.04 *ppm*.

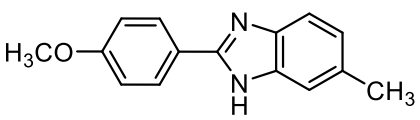
(17) 2-(Furan-2-yl)-1*H*-benzimidazole (**17c**):



The title compound **17c** was obtained following the general method for oxidative coupling using commercially available furan-2-ylmethanamine, **17a** and 1,2-diaminobenzene, **1b** affording the title compound as brown solid, compound data matches those previously reported.[7]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 7.90 (s, 1H), 7.56-7.54 (m, 2H), 7.20-7.19 (m, 3H), 6.71-6.70 (m, 1H) *ppm.*
^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 145.56, 144.89, 143.85, 122.59, 115.26, 112.60, 110.86 *ppm*.

(18) 2-(4-methoxyphenyl)-6-methyl-1*H*-benzimidazole (**18c**):

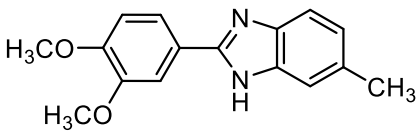


The title compound **18c** was obtained following the general method for oxidative coupling using commercially available 4-methoxybenzylamine, **2a** and 4-methylbenzene-1,2-diamine, **2b** affording the title compound as white solid, compound data matches those previously reported.[9]

^{1}H NMR (500 MHz, $CDCl_3$, 24 °C): δ 8.57-8.55 (m, 1H), 7.55 (bs, 1H), 7.42-7.39 (m, 2H), 7.15-7.12 (m, 1H), 7.09-7.04 (m, 2H), 4.06 (s, 3H), 2.49 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (126 MHz, $CDCl_3$, 24 °C): δ 156.85, 149.68, 131.16, 130.22, 124.27, 121.87, 118.13, 111.61, 56.09, 21.86 *ppm*.

(19) 2-(3,4-dimethoxyphenyl)-6-methyl-1*H*-benzimidazole (**19c**):

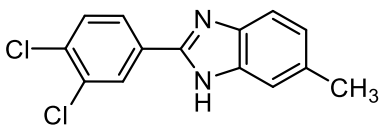


The title compound **19c** was obtained following the general method for oxidative coupling using commercially available 3,4-dimethoxybenzylamine, **19a** and 4-methylbenzene-1,2-diamine, **2b** affording the title compound as yellow solid, compound data matches those previously reported.[13]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 12.57 (bs, 1H), 7.74-7.70 (m, 2H), 7.44-7.34 (m, 2H),7.12-7.10 (m, 1H), 6.99-6.98 (m, 1H), 3.87 (s, 3H), 3.83 (s, 3H), 2.42 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 150.13, 148.88, 122.92, 119.10, 111.80, 109.67, 55.59, 55.57, 21.32 *ppm*.

(20) 2-(3,4-dichlorophenyl)-6-methyl-1*H*-benzo[d]imidazole (**20c**):



The title compound **20c** was obtained following the general method for oxidative coupling using 3,4-dichlorobenzylamine, **9a** and 4-methylbenzene-1,2-diamine, **2b** affording the title compound as white solid.[14]

^{1}H NMR (500 MHz, DMSO-d_6, 24 °C): δ 12.88 (bs, 1H), 8.35 (s, 1H), 8.11-8.10 (m, 1H), 7.78-7.76 (m, 1H), 7.53-7.32 (m, 2H), 7.03 (m, 1H), 2.42 (s, 3H) *ppm.*

^{13}C {^{1}H} NMR (126 MHz, DMSO-d_6, 24 °C): δ 148.71, 148.34, 144.00, 141.78, 135. 27, 131.77, 131.18, 130.85, 127.76, 126.24, 123.66, 118.68, 111.18, 21.34 *ppm*.

S17. NMR (^{1}H and ^{13}C) spectra of benzimidazoles

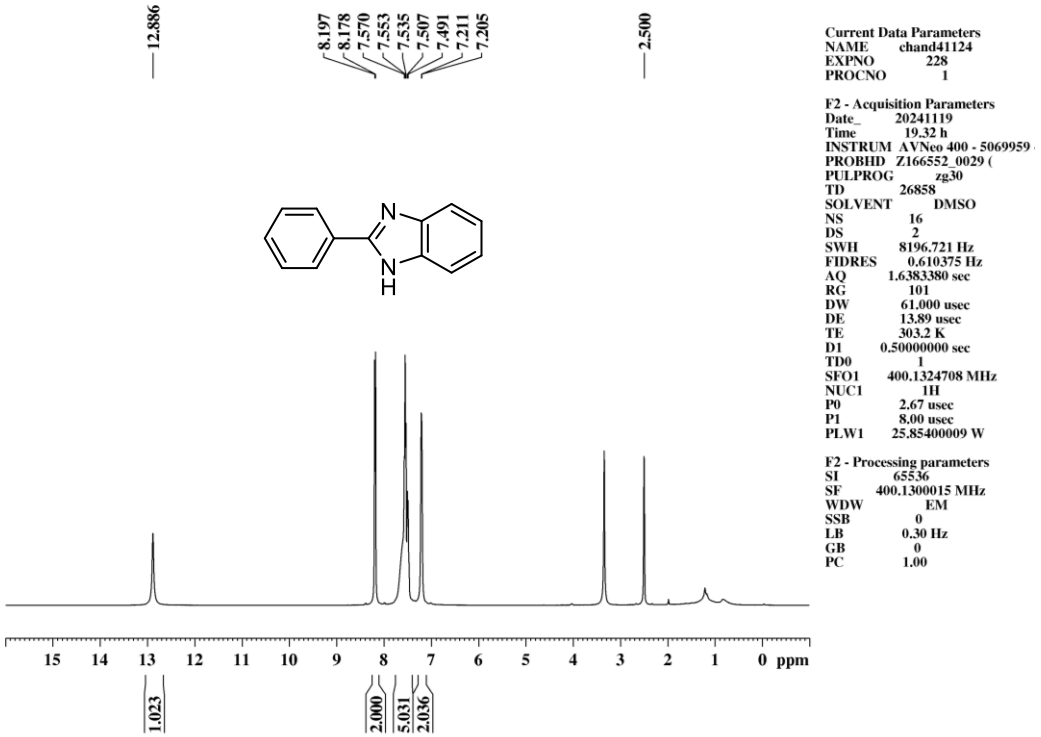


Figure S21 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-phenyl-1*H*-benzimidazole (**1c**).

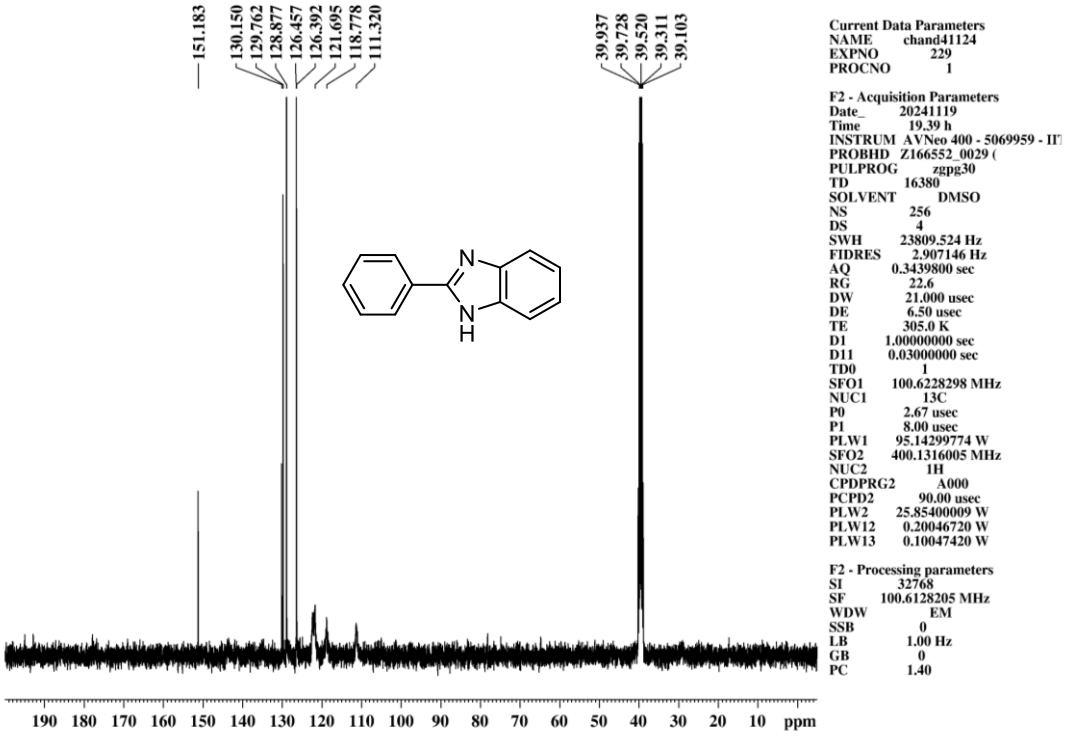


Figure S22 ^{1}H NMR spectrum (101 MHz, DMSO-d_6) of 2-phenyl-1*H*-benzimidazole (**1c**).

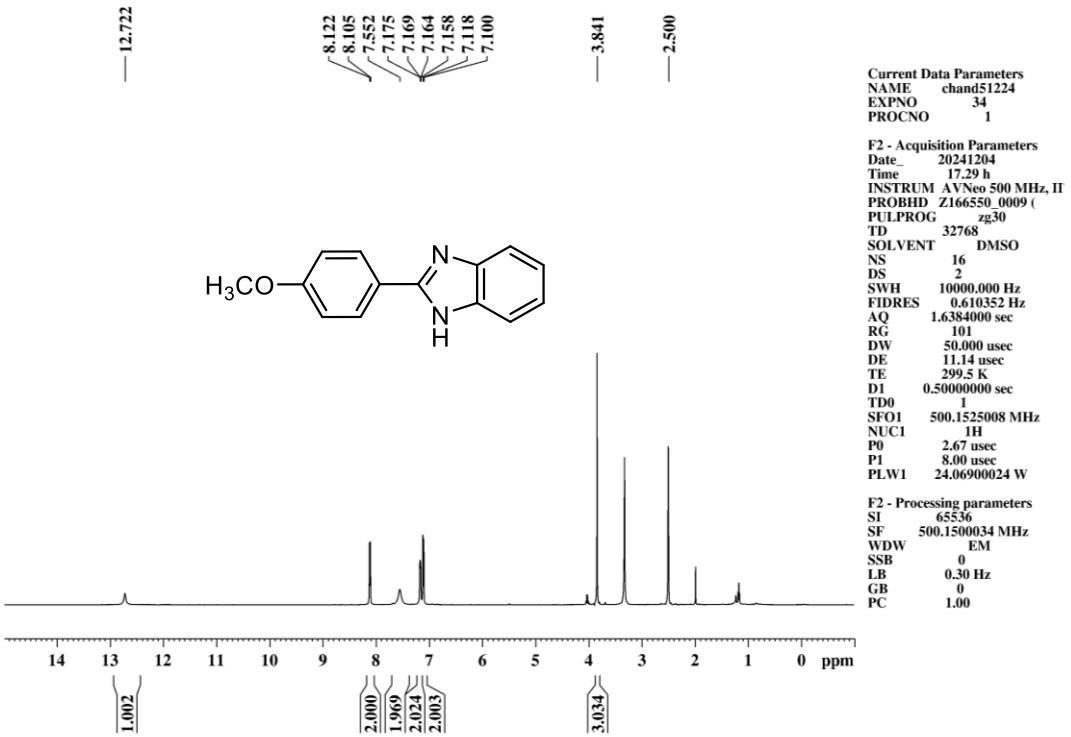


Figure S23 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(4-methoxyphenyl)-1*H*-benzimidazole (**2c**).

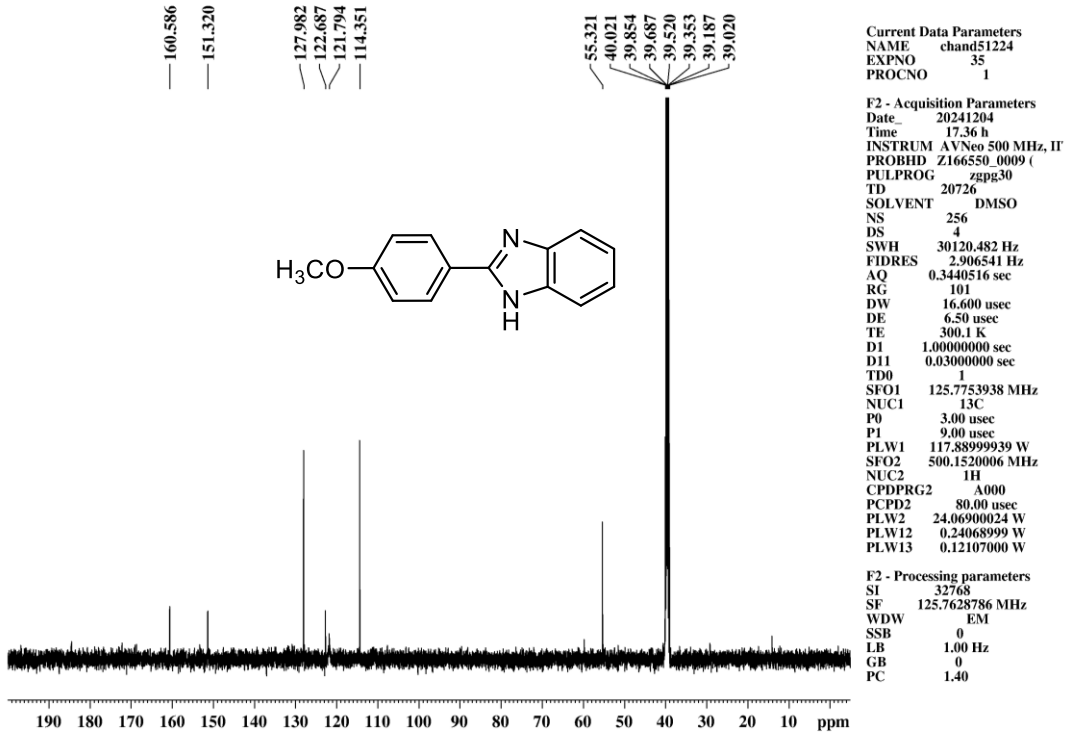


Figure S24 ^{1}H NMR spectrum (126 MHz, DMSO-d_6) of 2-(4-methoxyphenyl)-1*H*-benzimidazole (**2c**).

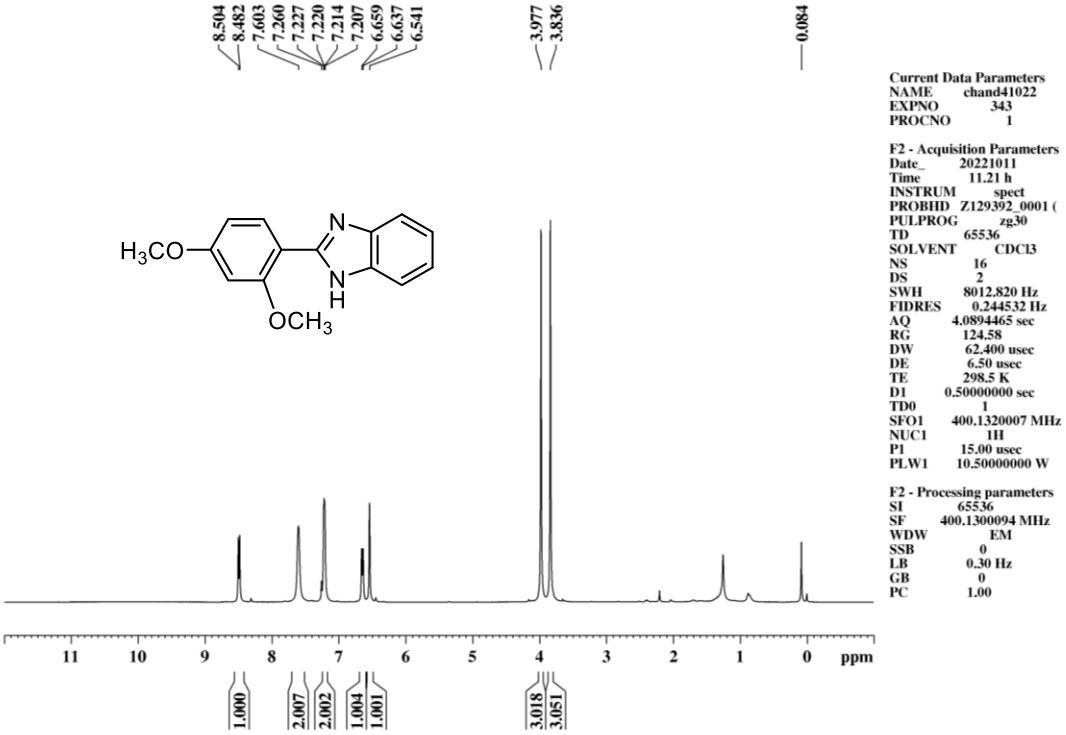


Figure S25 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(2,4-dimethoxyphenyl)-1*H*-benzimidazole (**3c**).

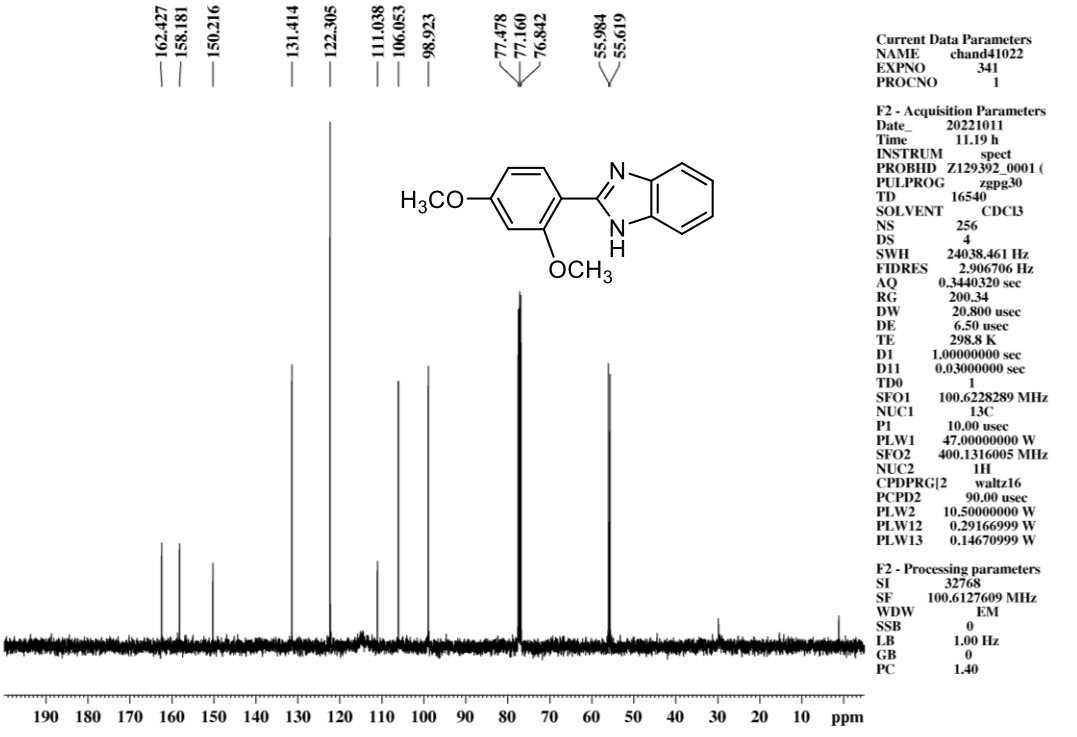


Figure S26 ^{1}H NMR spectrum (101 MHz, DMSO-d_6) of 2-(2,4-dimethoxyphenyl)-1*H*-benzimidazole (**3c**).

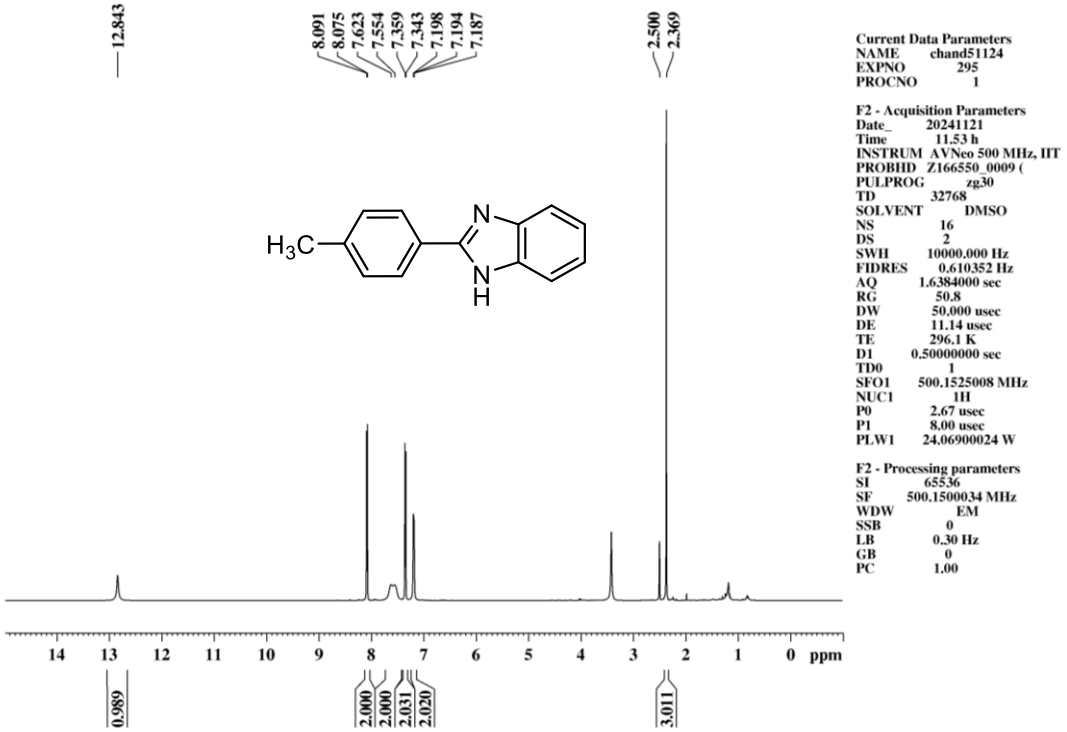


Figure S27 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(p-tolyl)-1*H*-benzimidazole (**4c**).

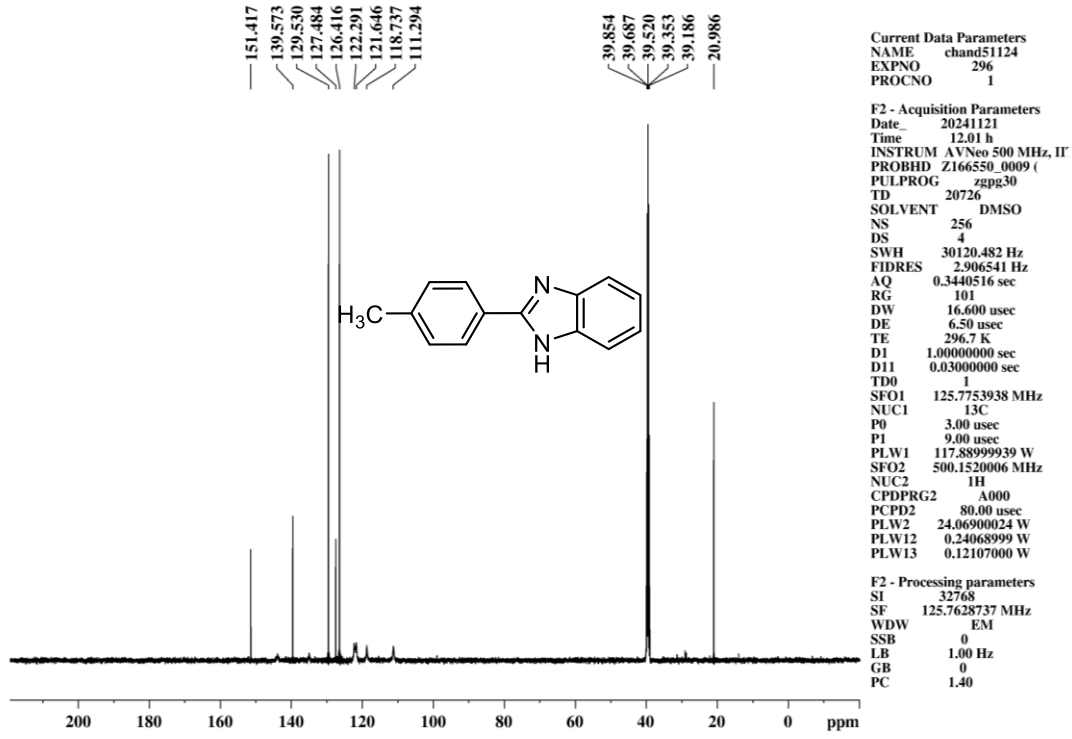


Figure S28 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(p-tolyl)-1*H*-benzimidazole (**4c**).

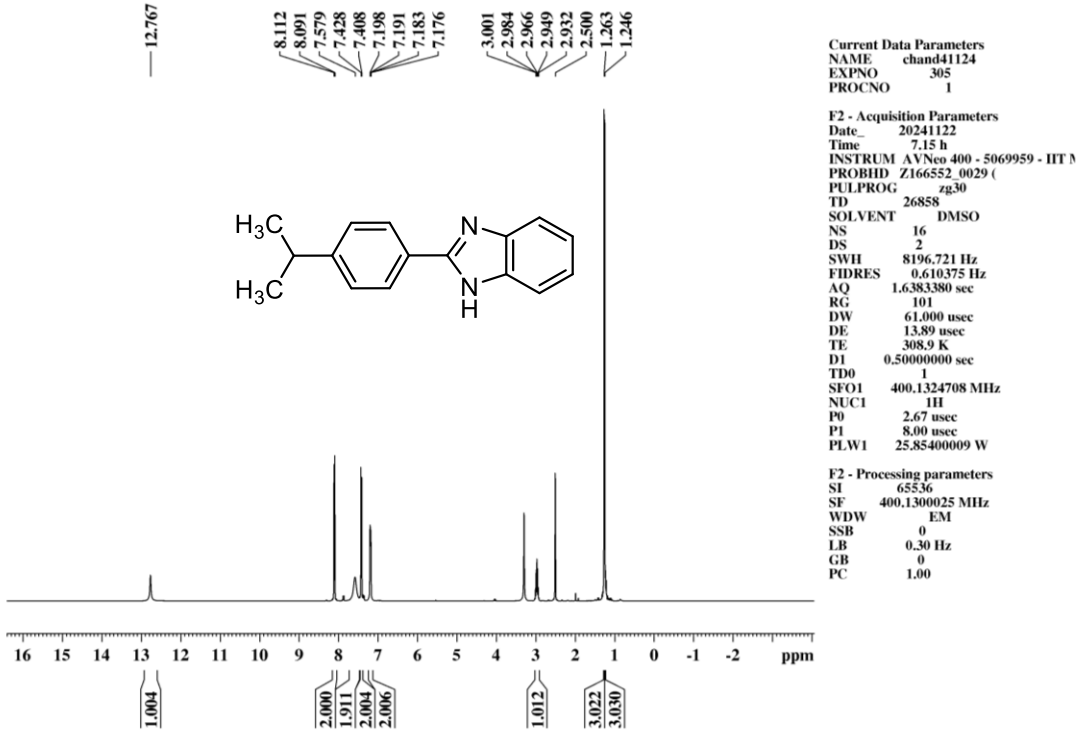


Figure S29 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(4-isopropylphenyl)-1*H*-benzimidazole (**5c**).

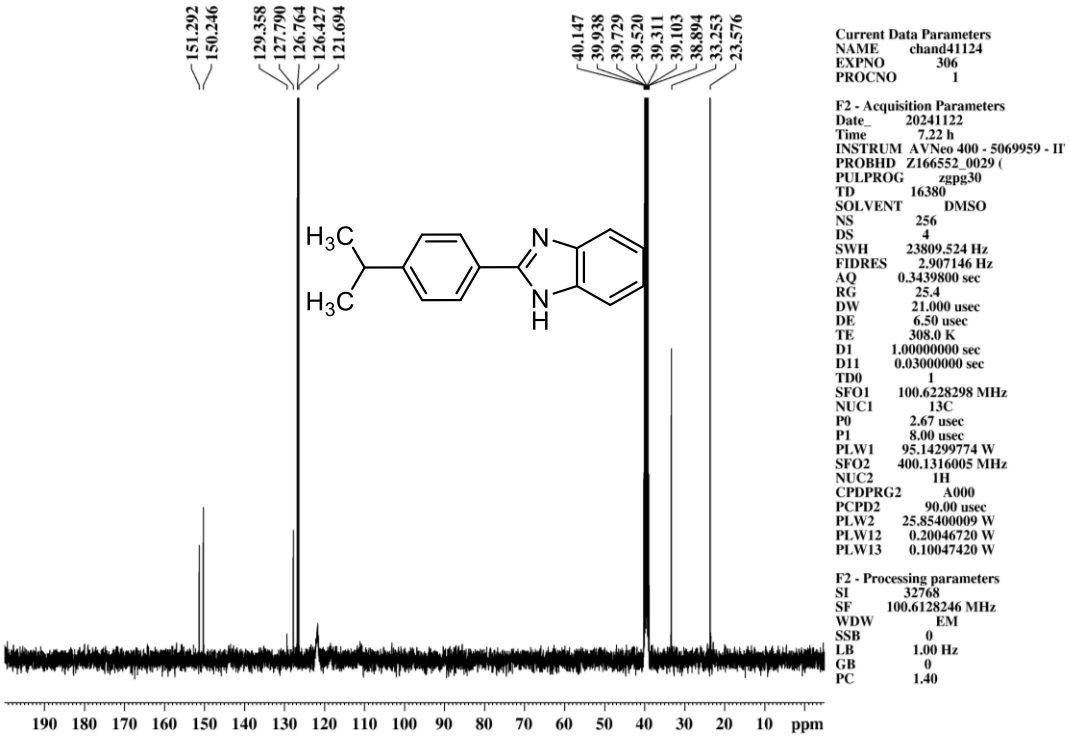


Figure S30 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(4-isopropylphenyl)-1*H*-benzimidazole (**5c**).

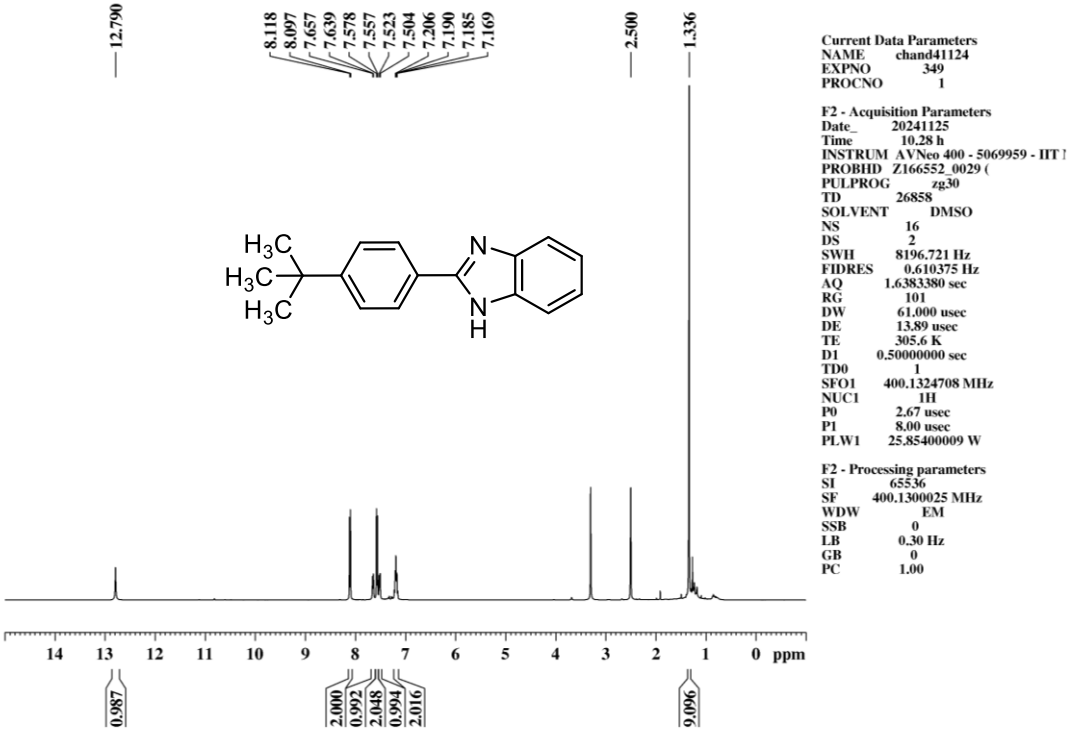


Figure S31 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(4-*tert*-butylphenyl)-1*H*-benzimidazole (**6c**).

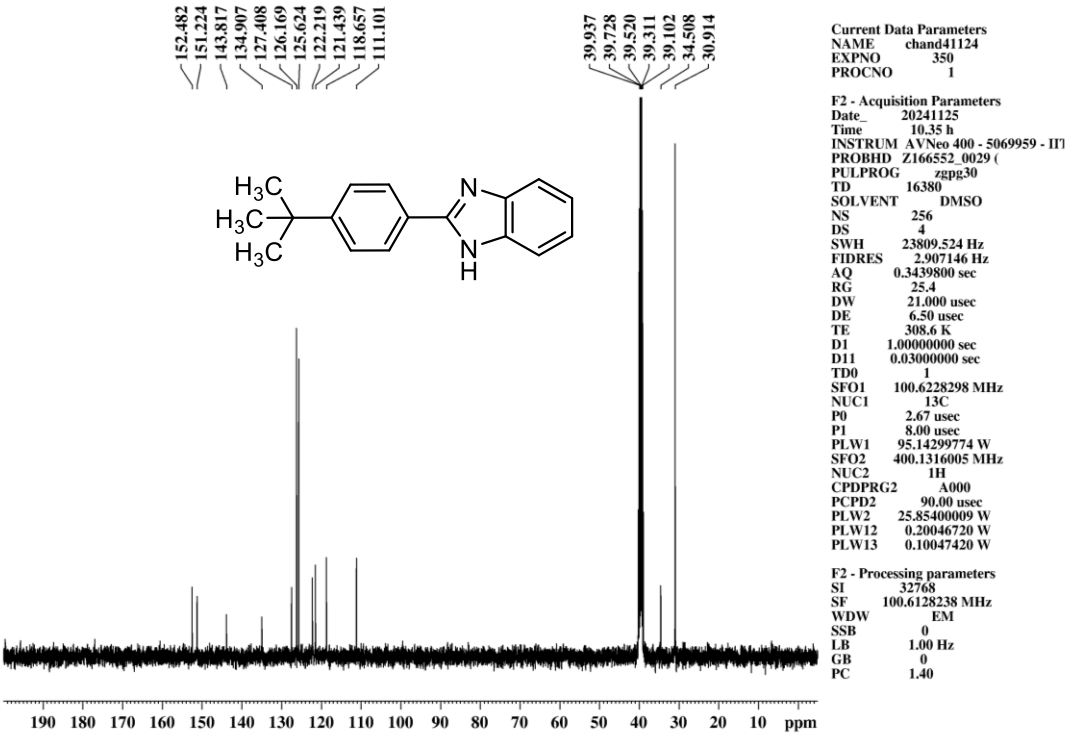


Figure S32 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(4-*tert*-butylphenyl)-1*H*-benzimidazole (**6c**).

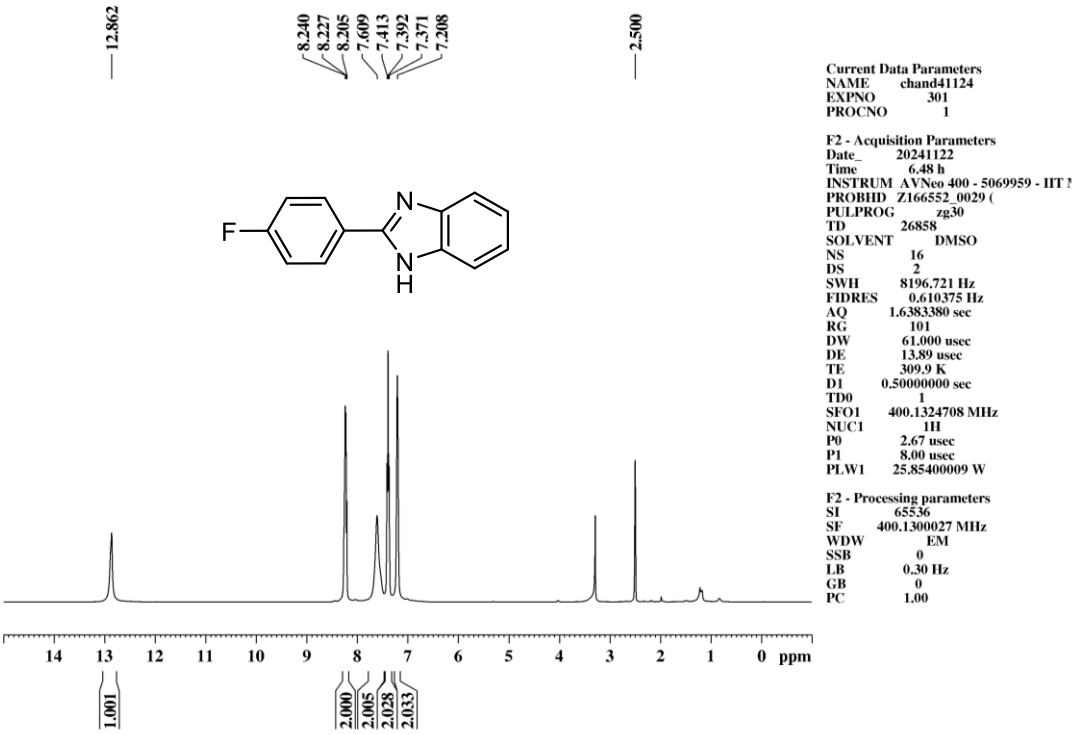


Figure S33 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(4-fluorophenyl)-1*H*-benzimidazole (**7c).**

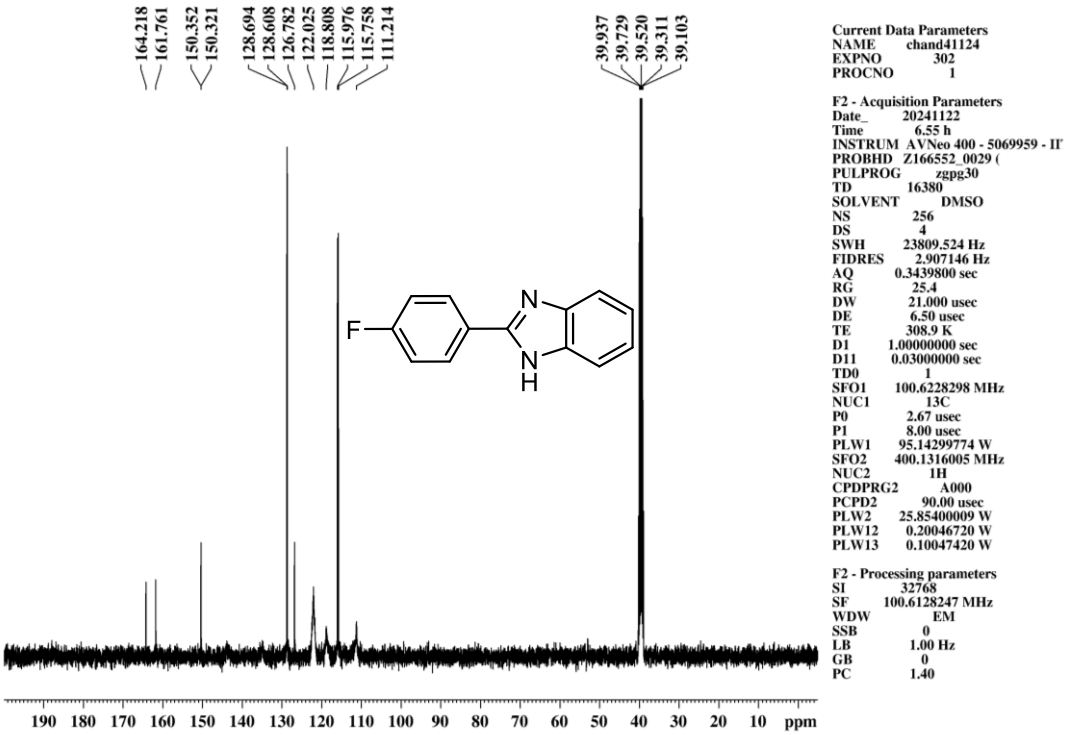


Figure S34 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(4-fluorophenyl)-1*H*-benzimidazole (**7c).**

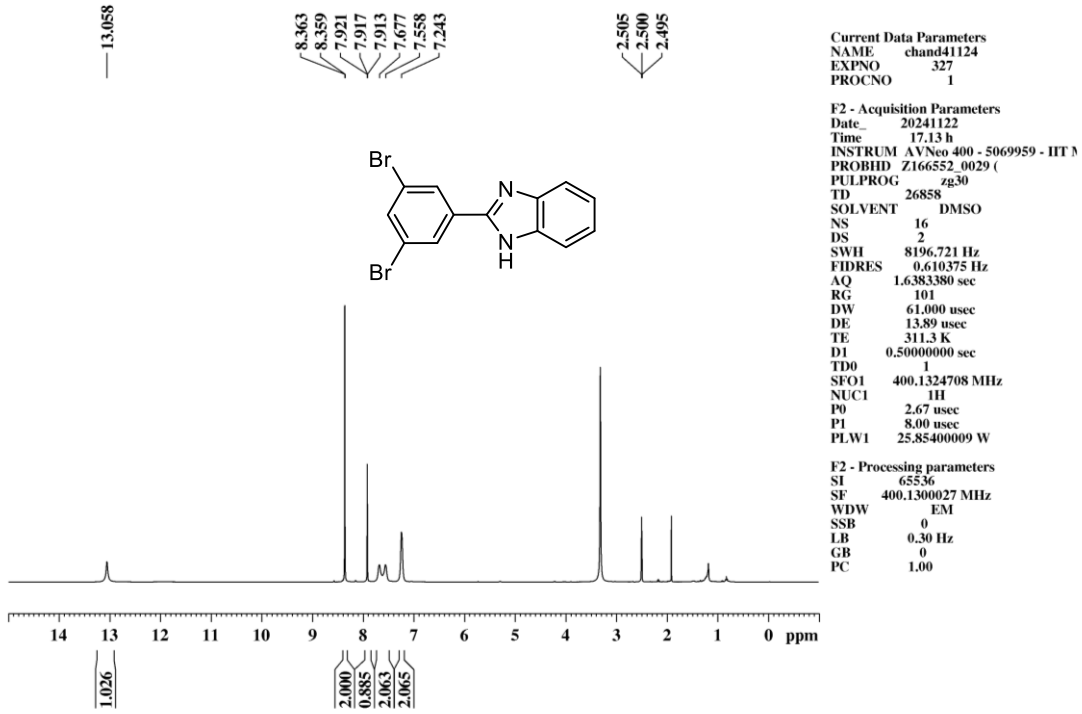


Figure S35 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(3,5-Dibromophenyl)-1*H*-benzimidazole (**8c**).

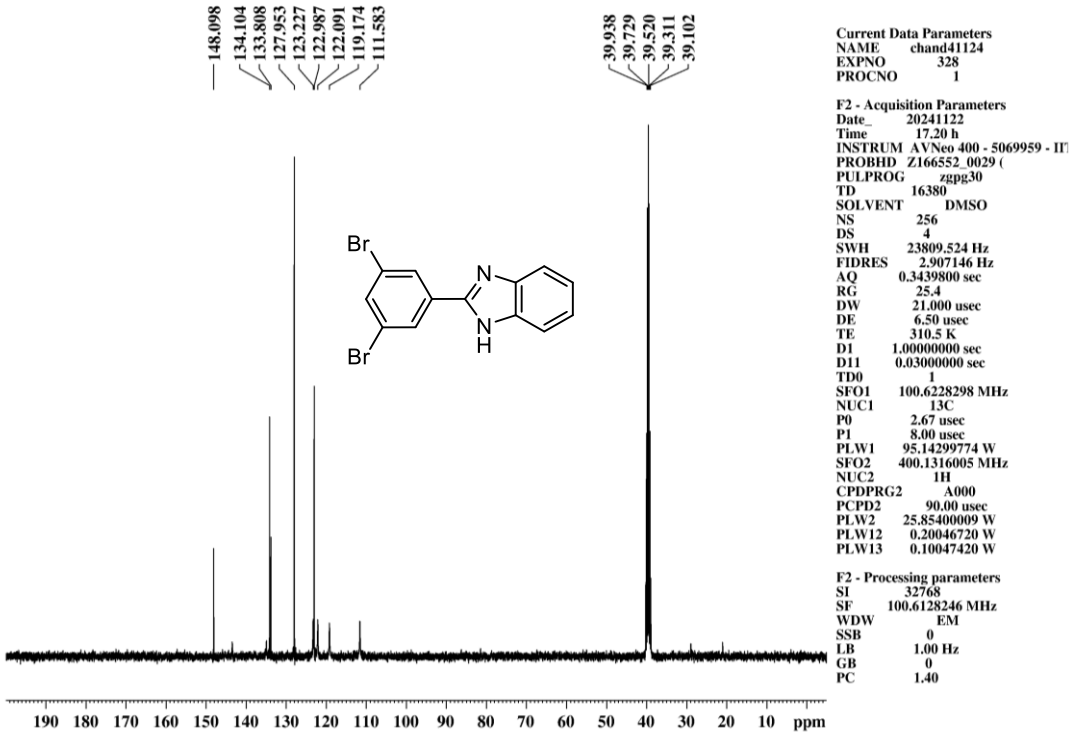


Figure S36 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(3,5-Dibromophenyl)-1*H*-benzimidazole (**8c**).

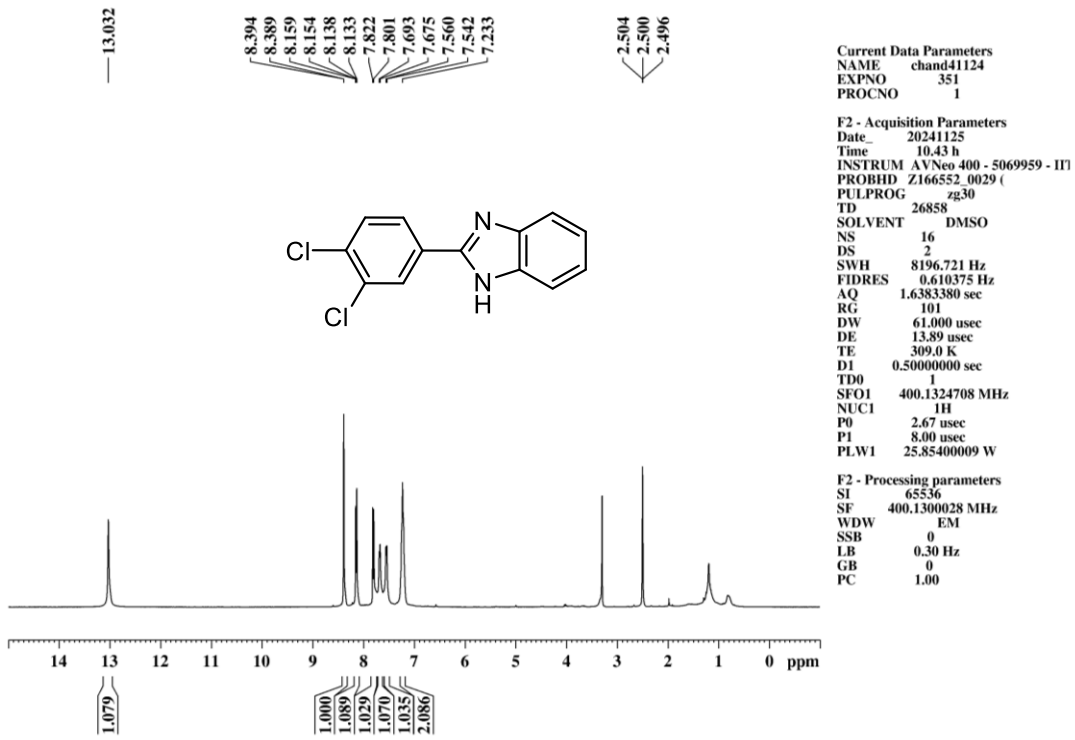


Figure S37 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(3,4-Dichlorophenyl)-1*H*-benzimidazole (**9c**).

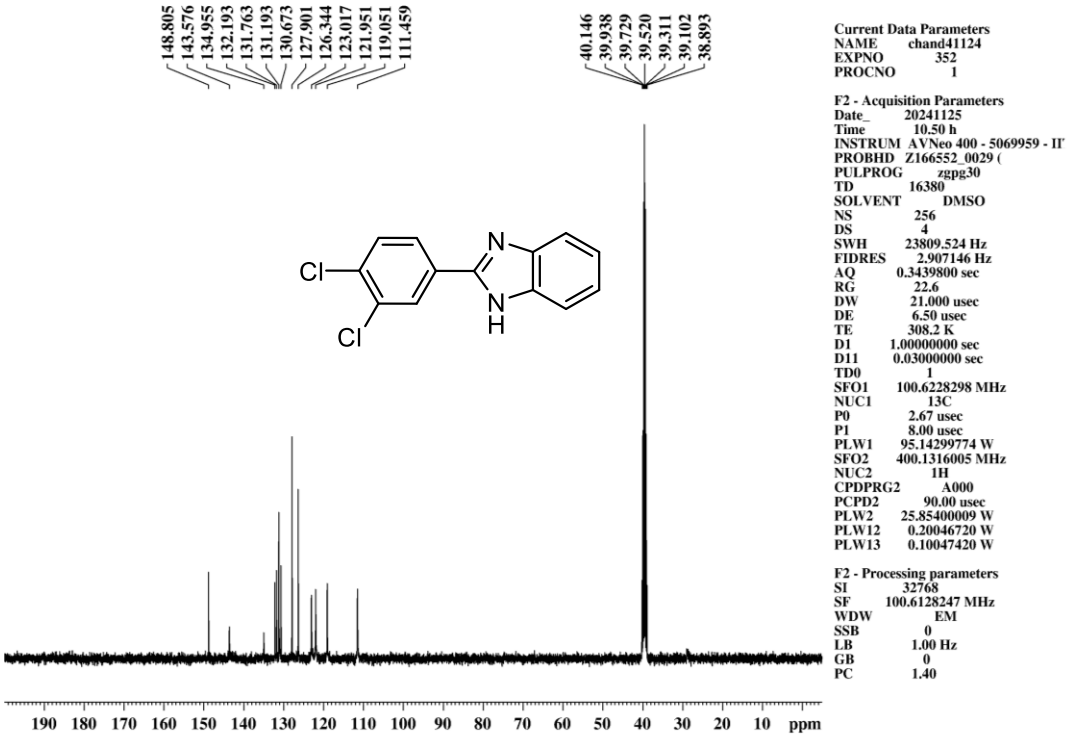


Figure S38 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(3,4-Dichlorophenyl)-1*H*-benzimidazole (**9c**).

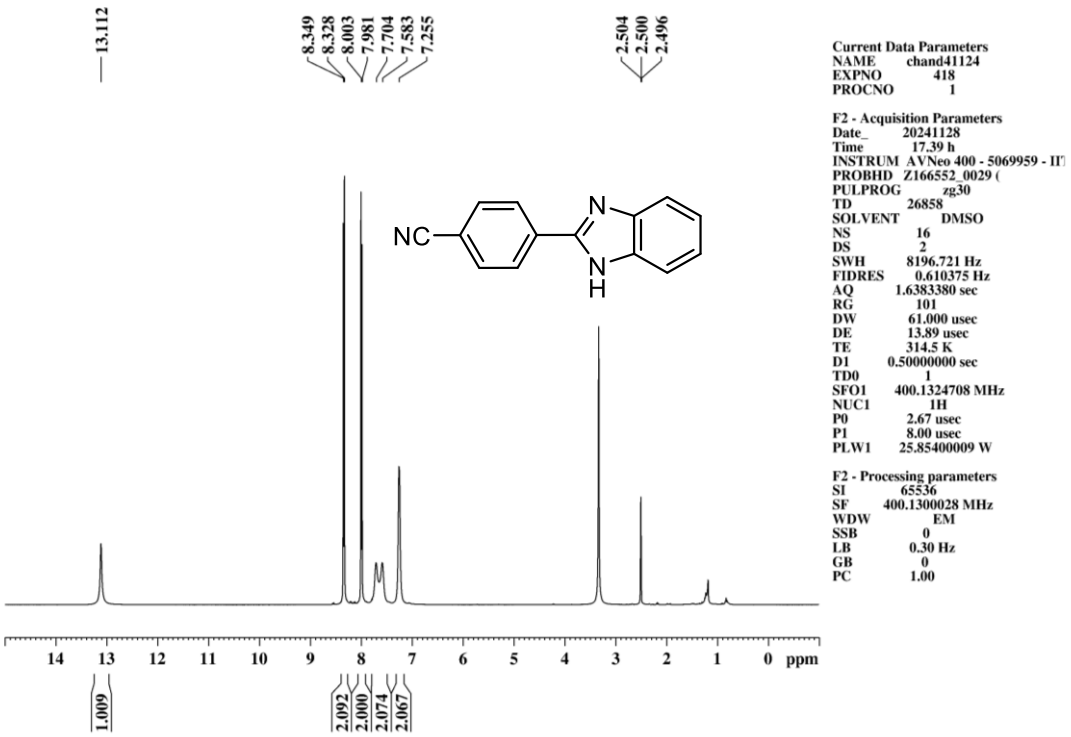


Figure S39 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 4-(1*H*-benzo[*d*]imidazol-2-yl)benzonitrile (**10c**).

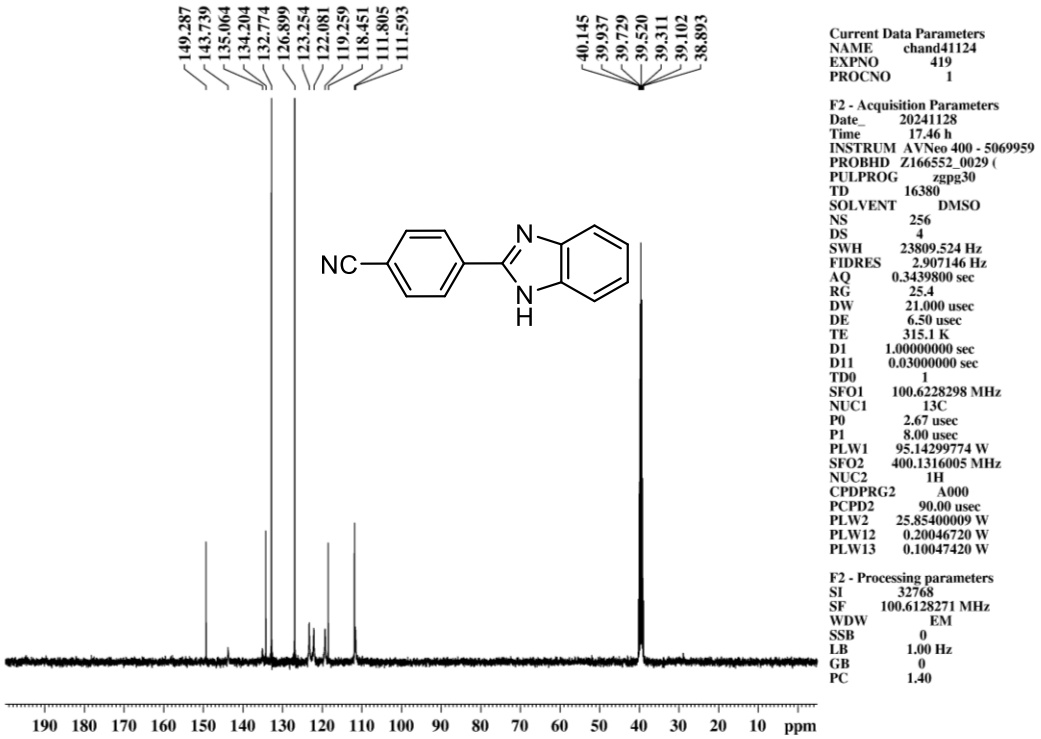


Figure S40 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 4-(1*H*-benzo[*d*]imidazol-2-yl)benzonitrile (**10c**).

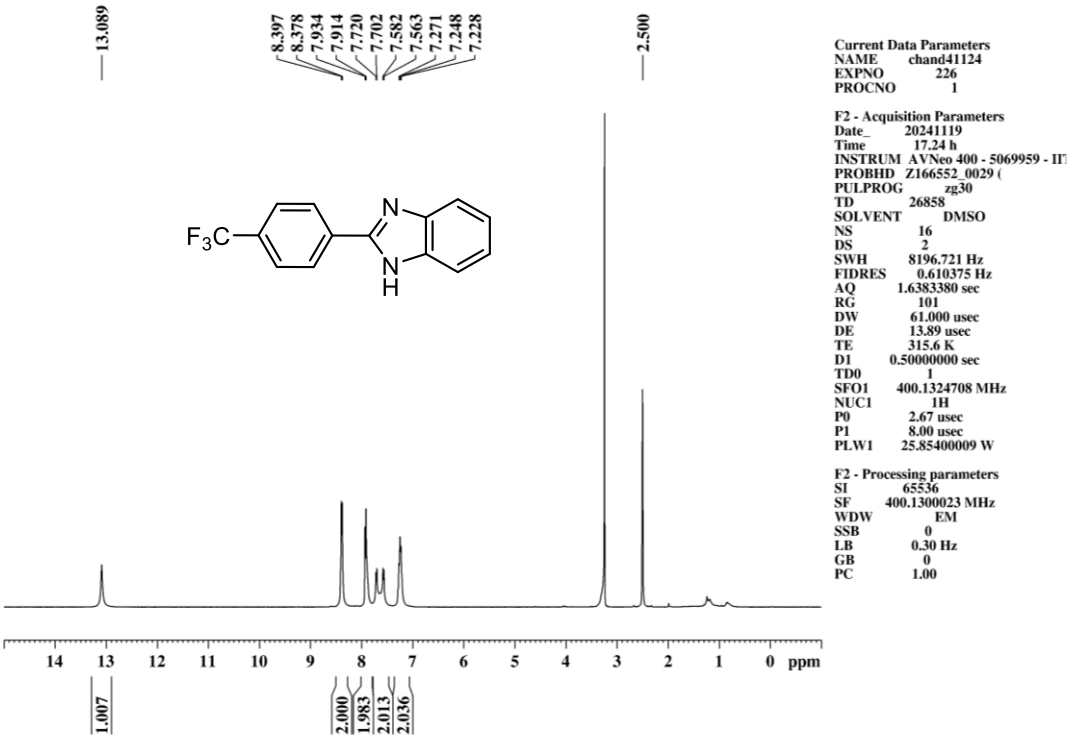


Figure S41 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(4-(trifluoromethyl)phenyl)-1*H*-benzimidazole (**11c**).

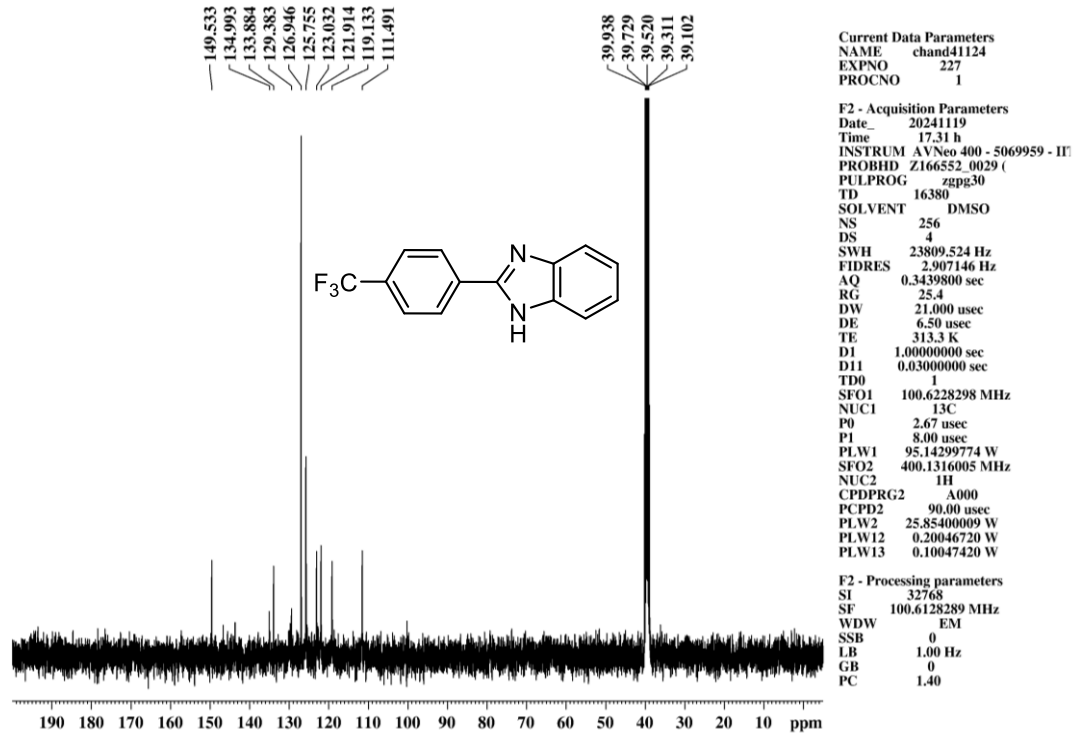


Figure S42 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(4-(trifluoromethyl)phenyl)-1*H*-benzimidazole (**11c**).

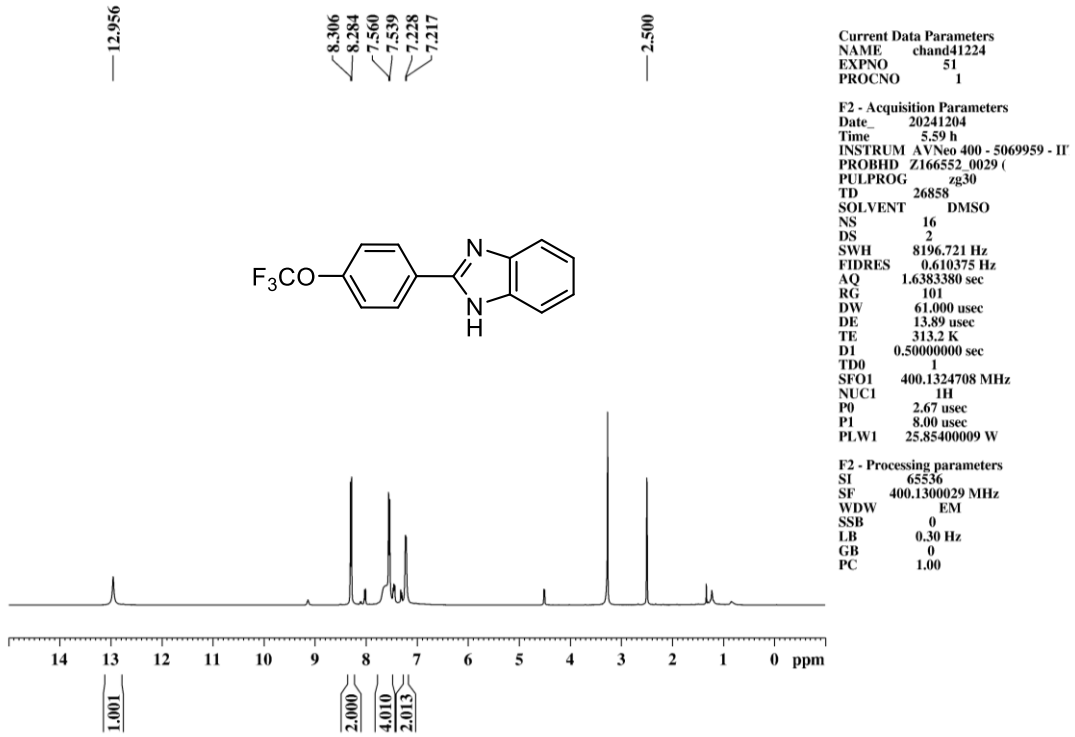


Figure S43 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(4-(trifluoromethoxy)phenyl)-1*H*-benzimidazole (**12c**).

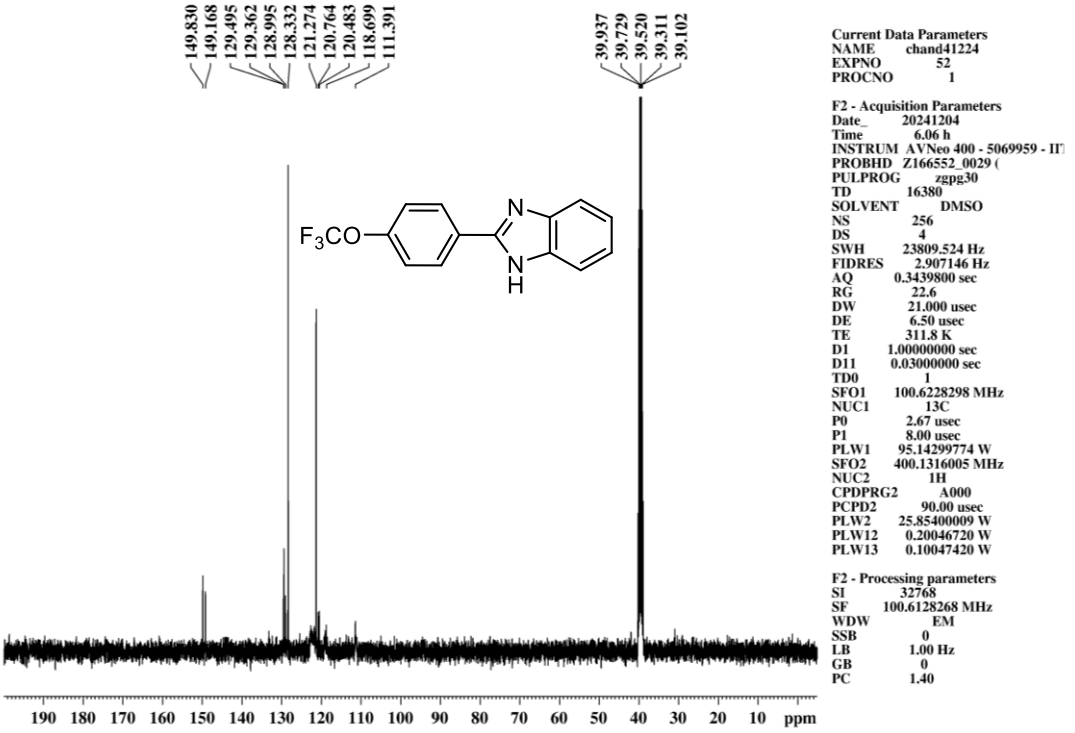


Figure S44 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(4-(trifluoromethoxy)phenyl)-1*H*-benzimidazole (**12c**).

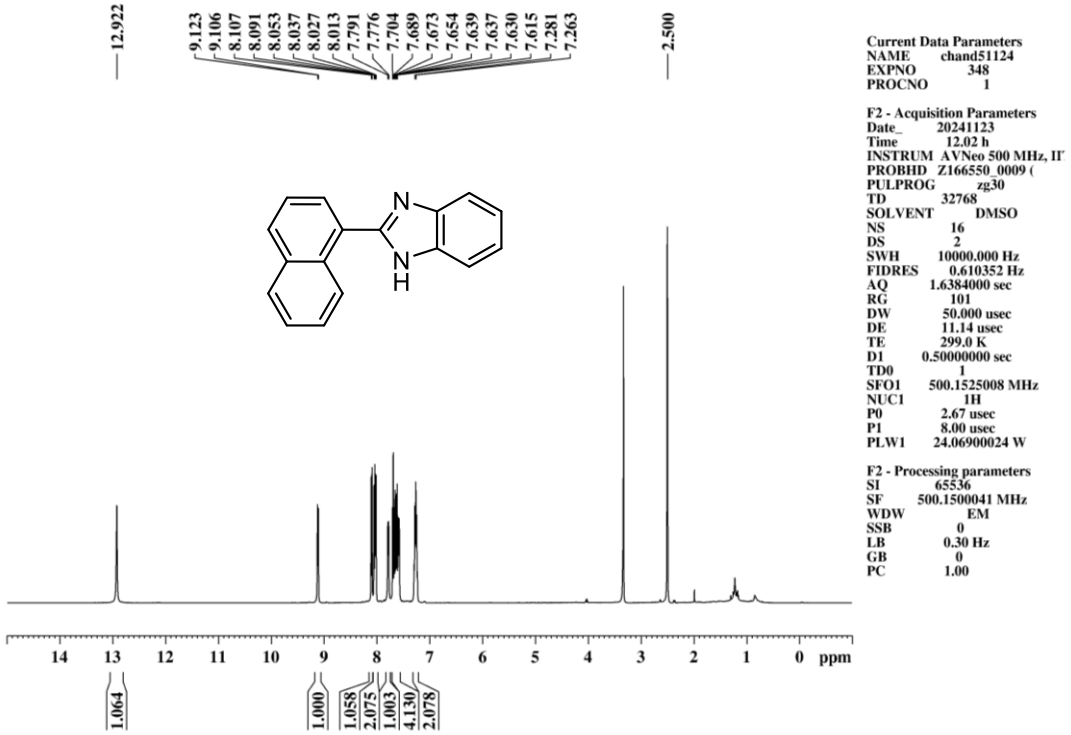


Figure S45 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(naphthalen-1-yl)-1*H*-benzimidazole (**13c**).

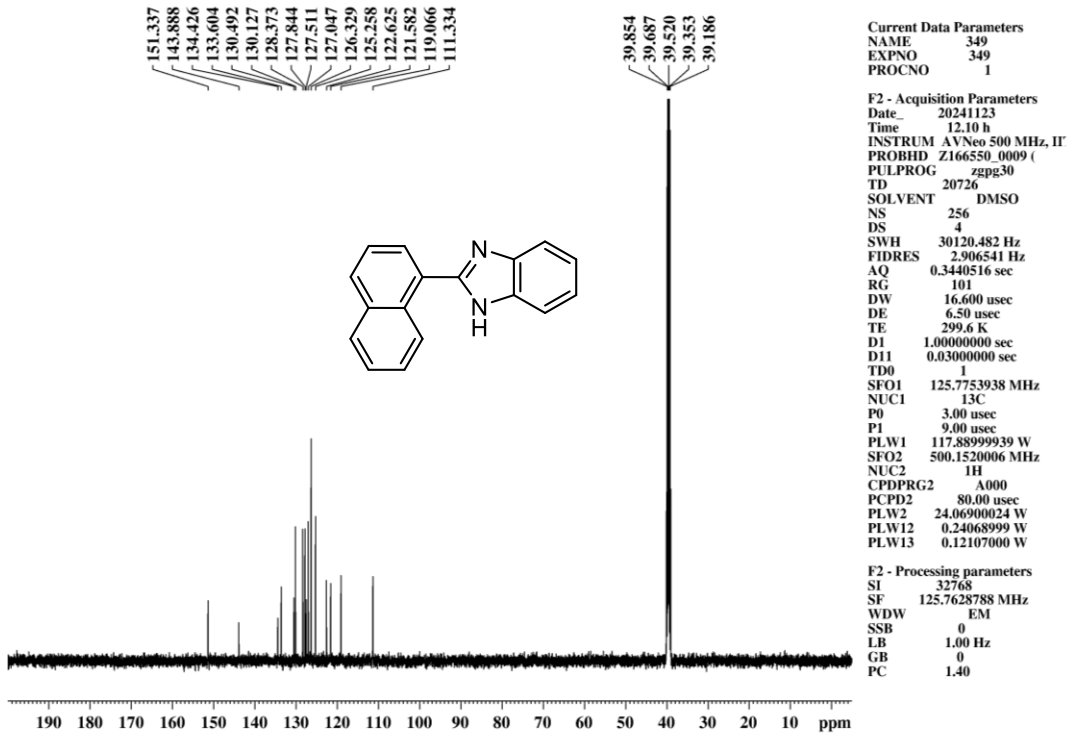


Figure S46 ^{13}C NMR spectrum (126 MHz, DMSO-d_6) of 2-(naphthalen-1-yl)-1*H*-benzimidazole (**13c**).

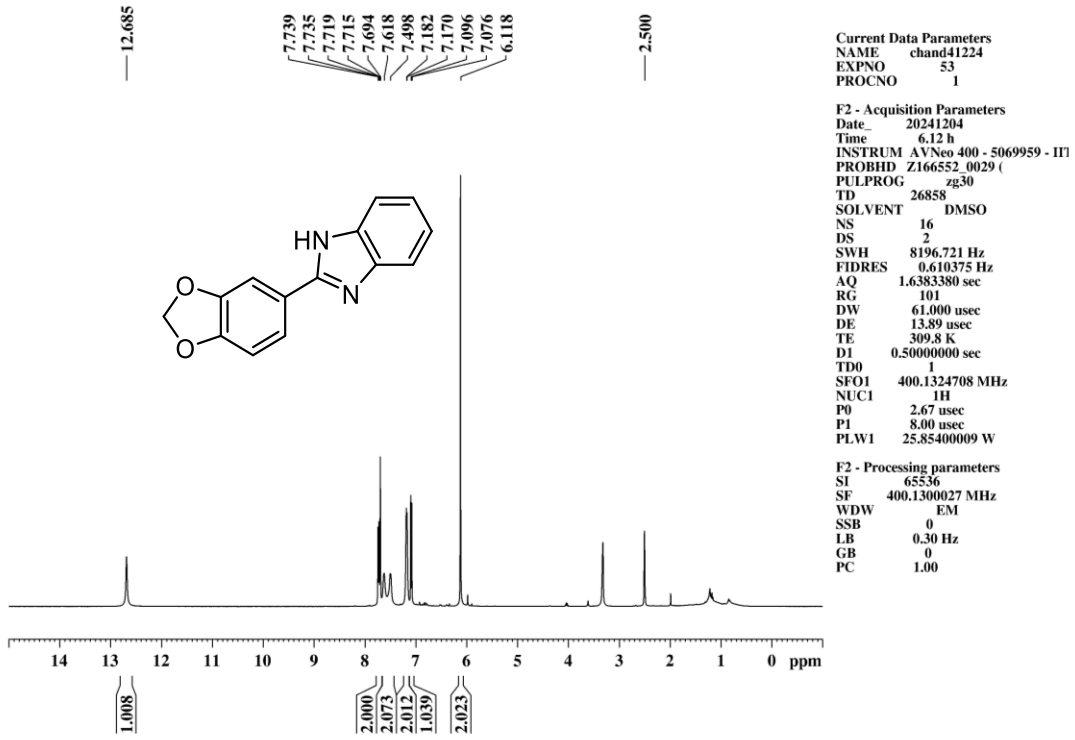


Figure S47 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(benzo[*d*][1,3]dioxol-5-yl)-1*H*-benzimidazole (**14c**).

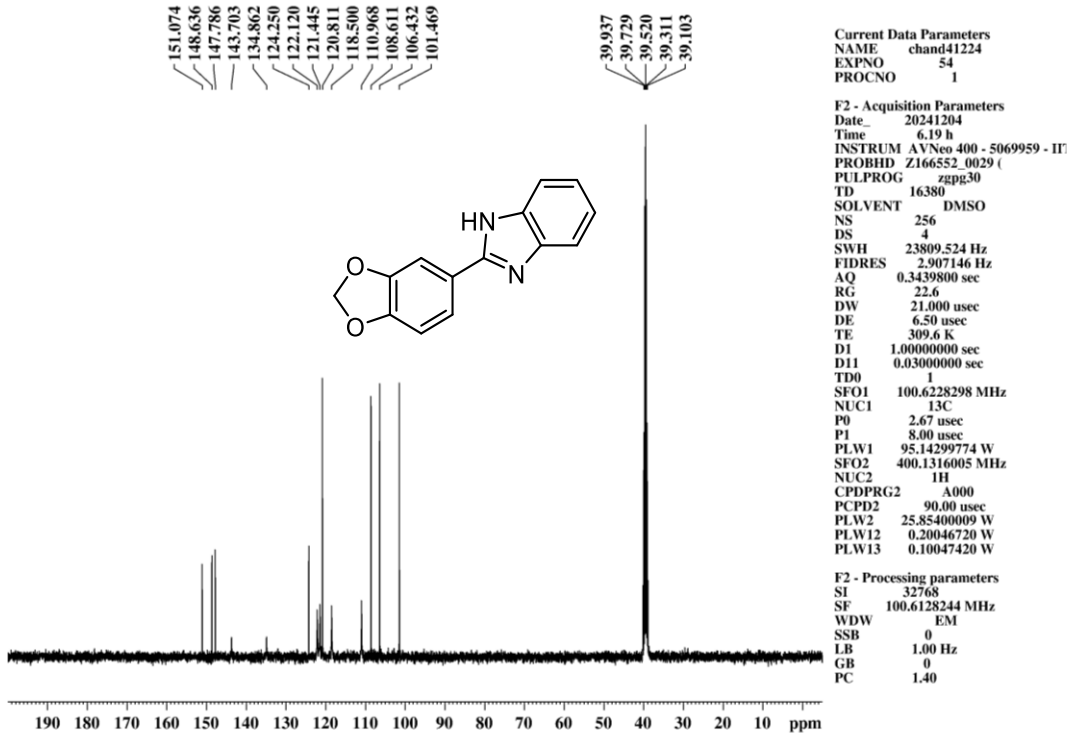


Figure S48 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(benzo[*d*][1,3]dioxol-5-yl)-1*H*-benzimidazole (**14c**).

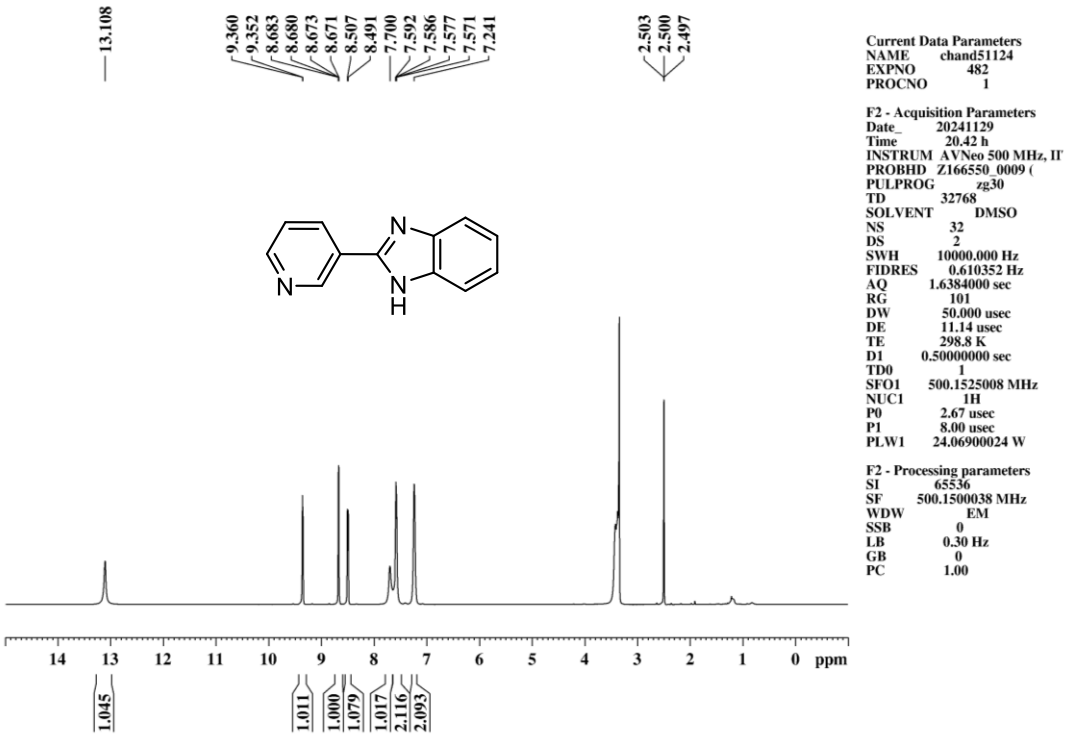


Figure S49 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(pyridin-2-yl)-1*H*-benzimidazole **(15c)**.

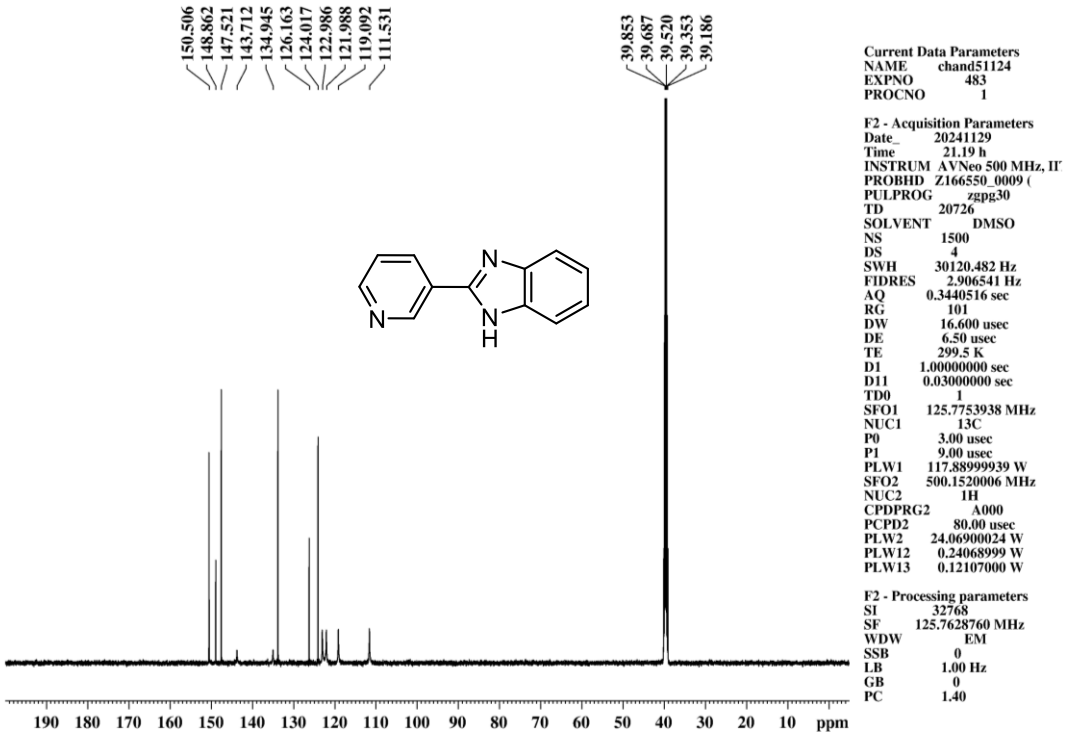


Figure S50 ^{13}C NMR spectrum (126 MHz, DMSO-d_6) of 2-(pyridin-2-yl)-1*H*-benzimidazole **(15c)**.

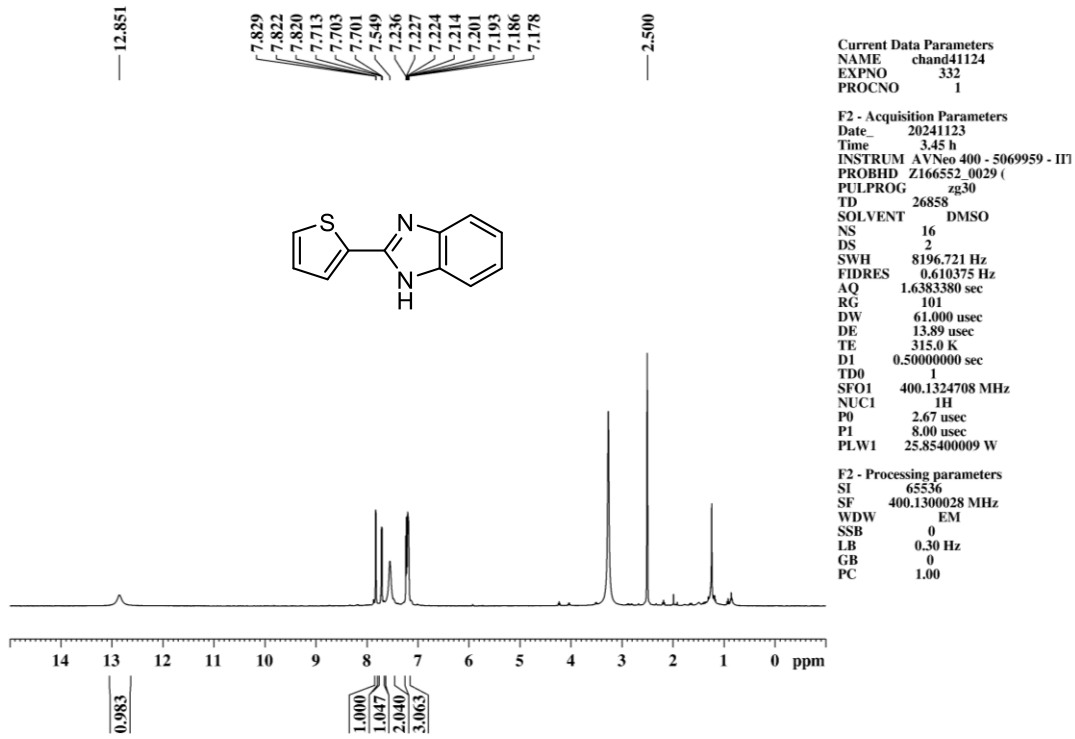


Figure S51 ^{1}H NMR spectrum (400 MHz, DMSO-d_6) of 2-(thiophen-2-yl)-1*H*-benzimidazole **(16c)**.

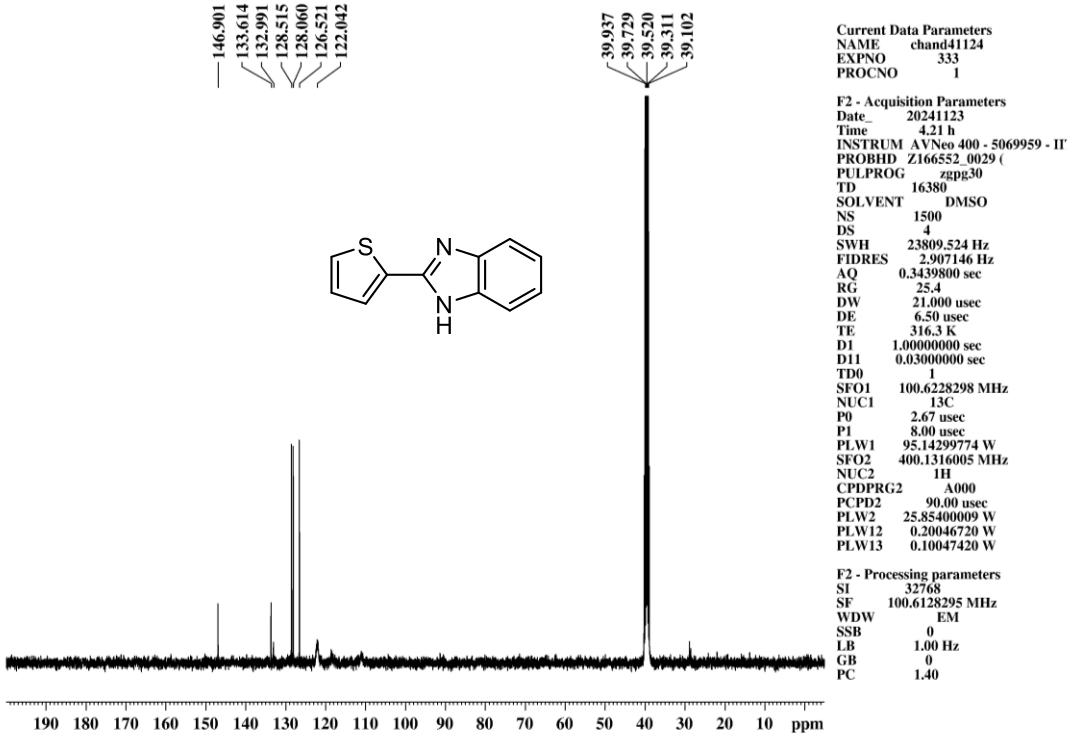


Figure S52 ^{13}C NMR spectrum (101 MHz, DMSO-d_6) of 2-(thiophen-2-yl)-1*H*-benzimidazole **(16c)**.

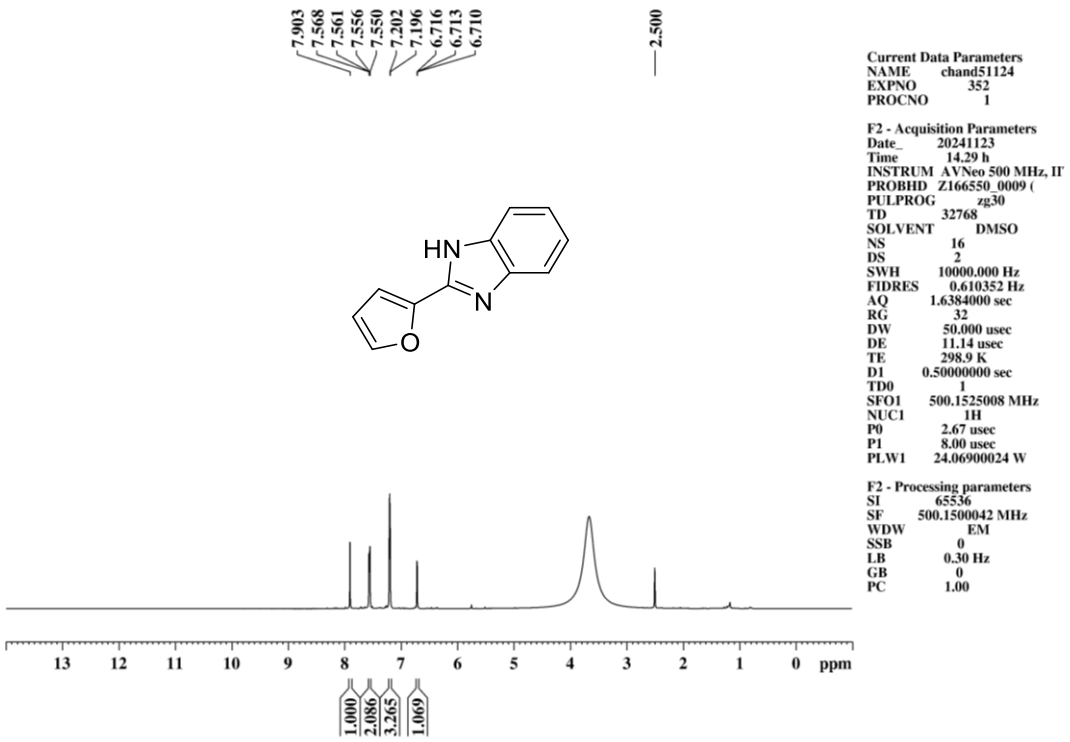


Figure S53 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(Furan-2-yl)-1*H*-benzimidazole (**17c**).

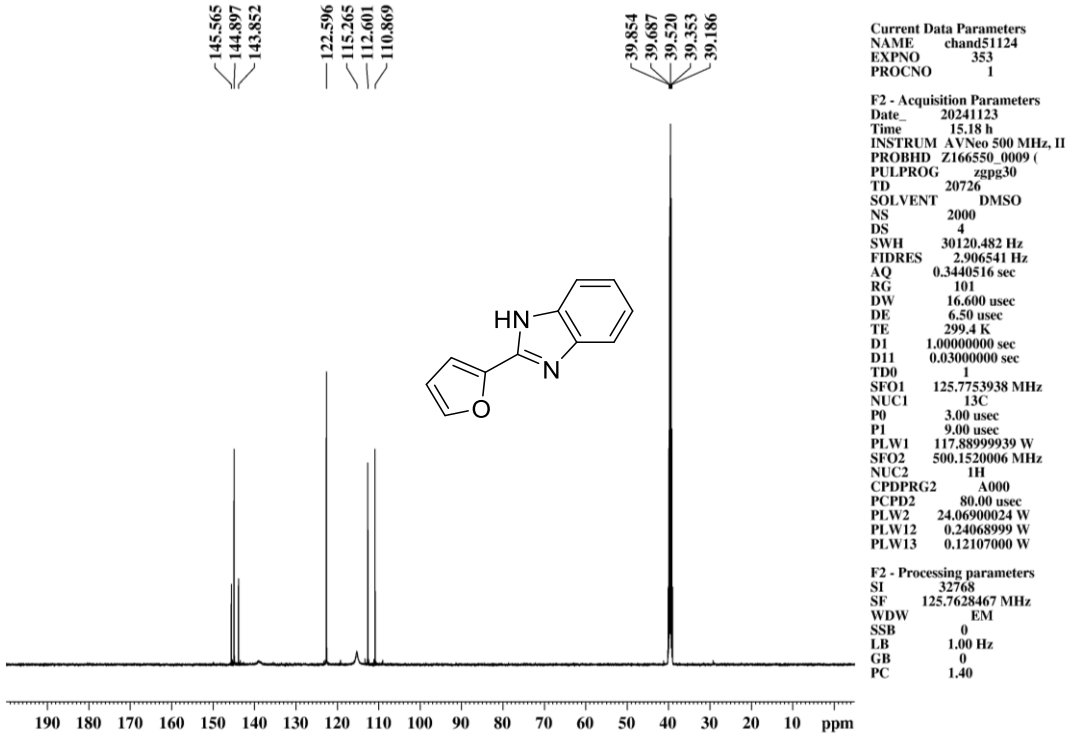


Figure S54 ^{13}C NMR spectrum (126 MHz, DMSO-d_6) of 2-(Furan-2-yl)-1*H*-benzimidazole (**17c**).

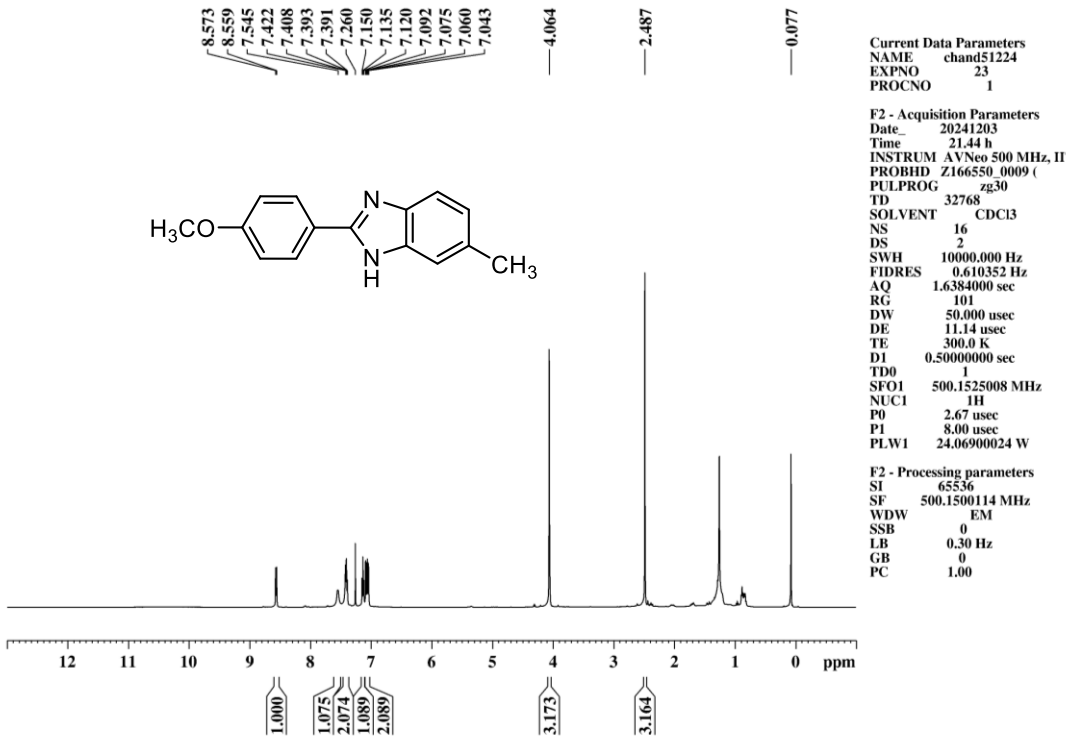


Figure S55 ^{1}H NMR spectrum (500 MHz, $CDCl_3$) of 2-(4-methoxyphenyl)-6-methyl-1*H*-benzimidazole (**18c**).

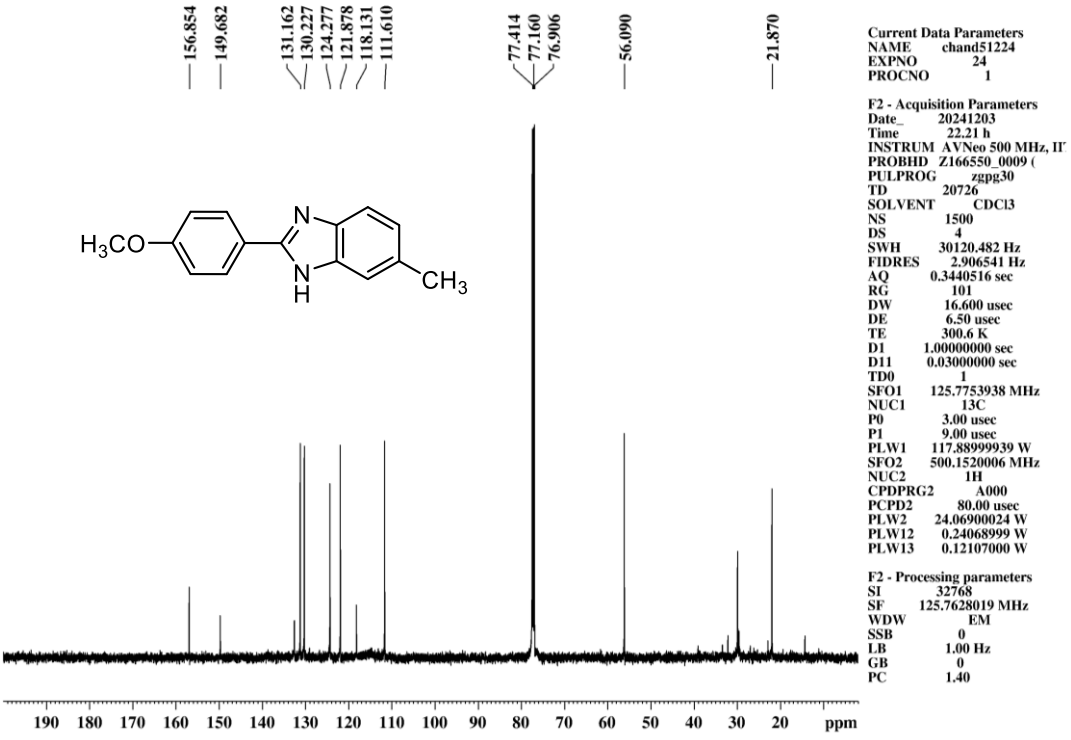


Figure S56 ^{13}C NMR spectrum (126 MHz, $CDCl_3$) of 2-(4-methoxyphenyl)-6-methyl-1*H*-benzimidazole (**18c**).

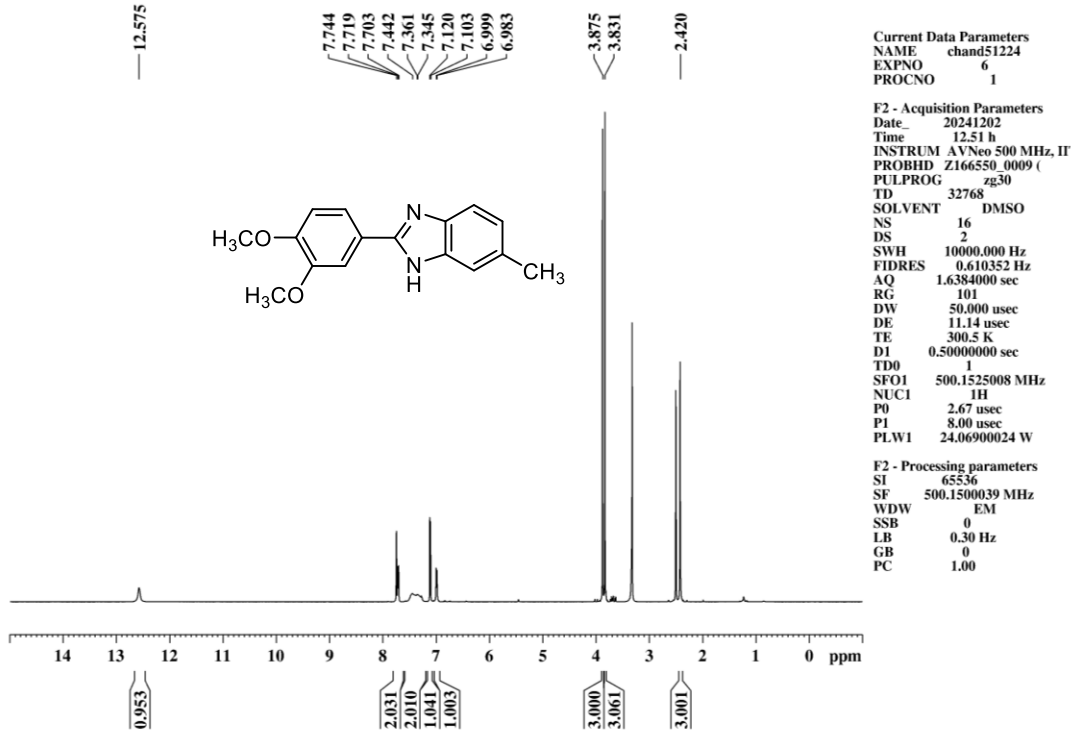


Figure S57 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(3,4-dimethoxyphenyl)-6-methyl-1*H*-benzimidazole (**19c**).

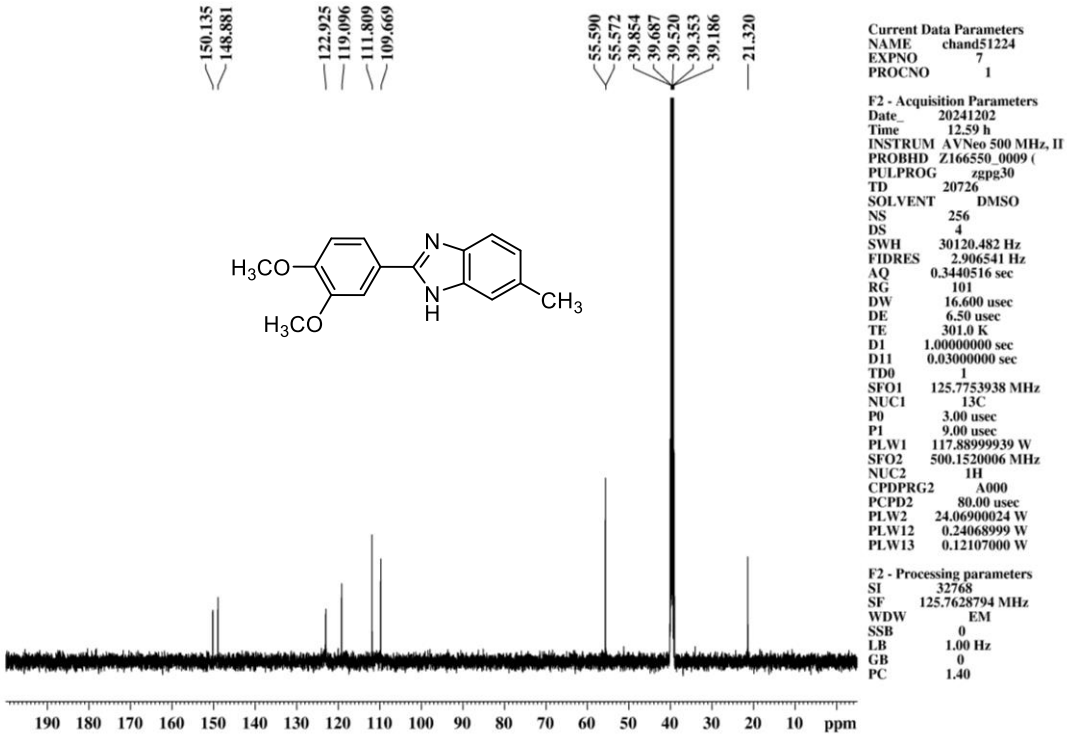


Figure S58 ^{13}C NMR spectrum (126 MHz, DMSO-d_6) of 2-(3,4-dimethoxyphenyl)-6-methyl-1*H*-benzimidazole (**19c**).

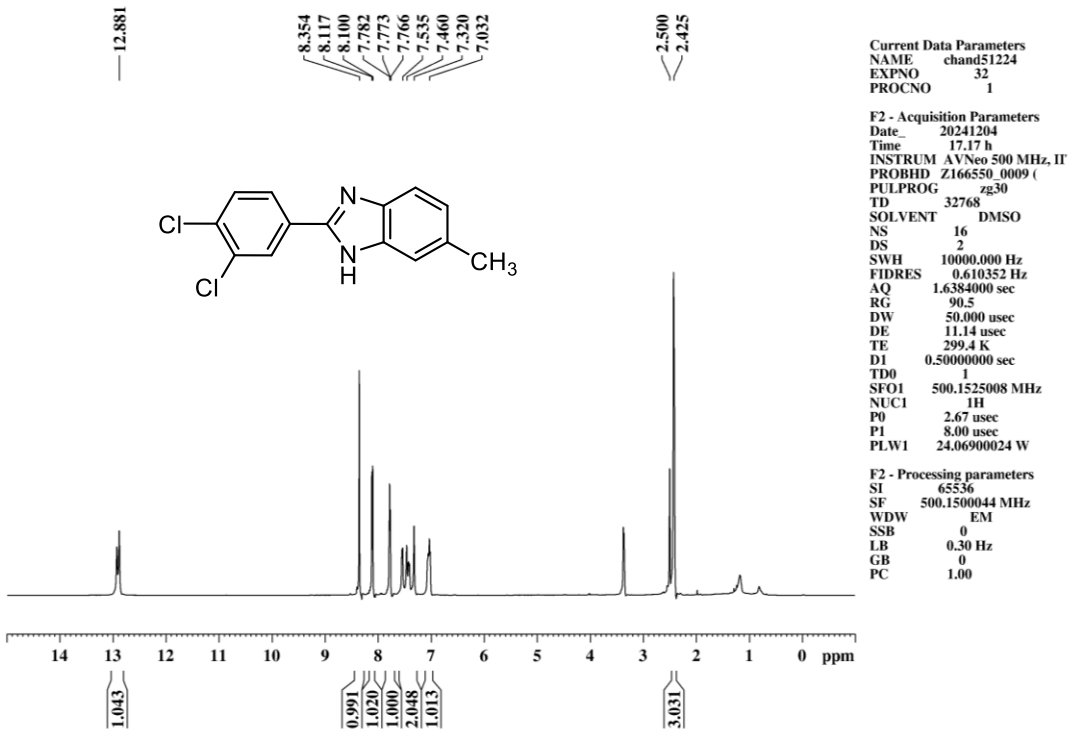


Figure S59 ^{1}H NMR spectrum (500 MHz, DMSO-d_6) of 2-(3,4-dichlorophenyl)-6-methyl-1*H*-benzimidazole (**20c**).

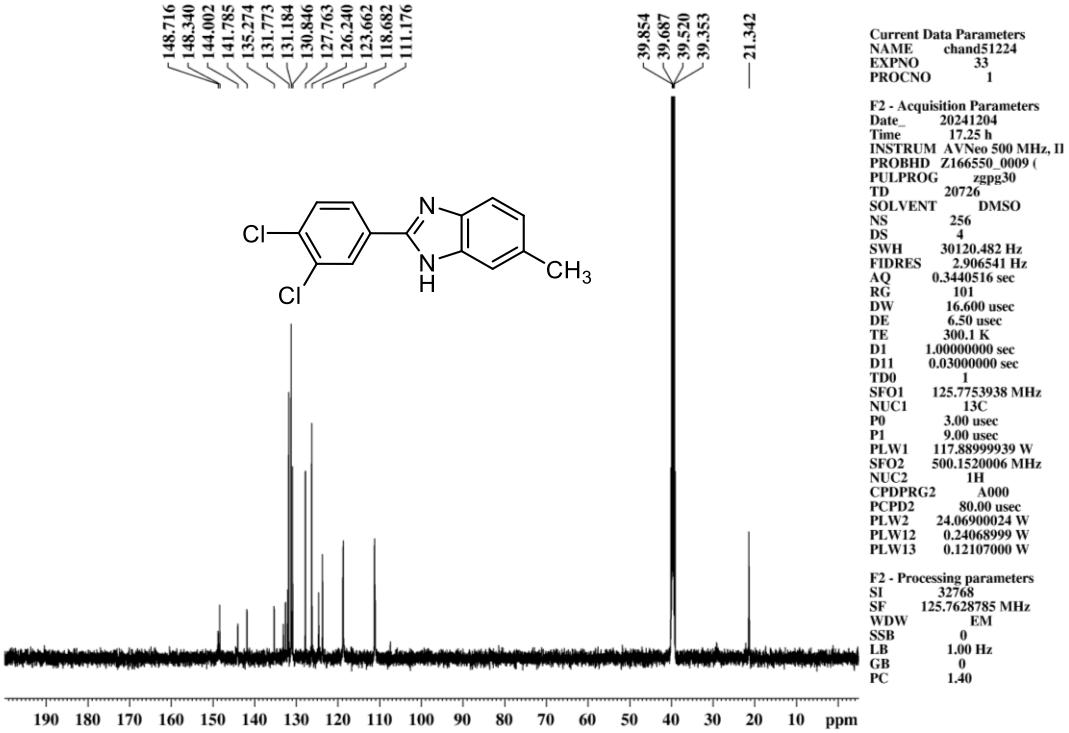


Figure S60 ^{13}C NMR spectrum (126 MHz, DMSO-d_6) of 2-(3,4-dichlorophenyl)-6-methyl-1*H*-benzimidazole (**20c**).

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