

Supporting Information

An efficient multicomponent synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazoles catalyzed by a magnetic nanoparticle supported Lewis acidic deep eutectic solvent.

Thanh Thi Nguyen, Ngoc-Phuong Thi Le, The Thai Nguyen and Phuong Hoang Tran*

*Department of Organic Chemistry, Faculty of Chemistry, University of Science, Vietnam National University, Ho Chi Minh City 721337, Vietnam.

Corresponding author: E-mail: thphuong@hcmus.edu.vn

Table of contents

Section S1. Chemicals and analytical Techniques	S2
Section S2. Synthesis of LADES@MNP catalyst	S3
Section S3. Spectral data of products	S4-S16
Section S4. Copies of ^1H and ^{13}C NMR spectra of all products	S17- S45
Section S5. References	S46 – S47

Section S1. Chemicals and analytical Techniques

Materials

Imidazole (99%), iron(II) chloride hexahydrate (99%), iron(II) sulfate heptahydrate (99%), potassium hydroxide (85%), benzyl ($\geq 99\%$), aniline ($\geq 99.5\%$), ammonium acetate ($\geq 98\%$) were purchased from Acros Organics. Zinc chloride ($\geq 98\%$), 3-(chloropropyl) triethoxysilane (95%), benzaldehyde (99%), 2-nitrobenzaldehyde (98%), 4-nitrobenzaldehyde (98%), 3-hydroxybenzaldehyde (99%), 4-hydroxybenzaldehyde (98%), 2-hydroxybenzaldehyde (97%), 4-florobenzaldehyde (98%), 4-methylbenzaldehyde (97%), 4-methoxybenzaldehyde (98%), 4-N,N-dimethylbenzaldehyde (99%), 4-chlorobenzaldehyde (97%), 4-t-butylbenzaldehyde (97%), 2-carboxybenzaldehyde (97%), 2-methyl-5-hydroxybenzaldehyde (97%) were obtained from Sigma-Aldrich Chemical Company. Ethyl acetate ($\geq 99.5\%$), petroleum ether ($\geq 99.5\%$), sodium sulfate anhydrous ($\geq 99\%$) were prepared from Xilong Chemical Co., Ltd. Ethanol ($\geq 99.5\%$) was obtained from Chemsol Co., Ltd. TLC (silica gel 60 F₂₅₄) layer was obtained from Merk.

Analytical Techniques

¹H and ¹³C nuclear magnetic resonance (NMR) spectra were recorded on a Bruker Advance II 500 MHz NMR spectrometer. Fourier Transform infrared (FT-IR) spectra were recorded from KBr pellets using a Bruker Vertex 70 system. Raman spectra were recorded on a Horiba Xplora One using a 532 nm argon ion laser. ICP-MS was recorded on a PerkinElmer 350X. Thermal gravimetric analysis (TGA) was performed on a TA Q500 thermal analysis system with the sample held in a platinum pan in a continuous airflow. TEM was recorder from JEOL JEM-1400 series 100kV Transmission Electron Microscope.

Section S2. Synthesis and characterization of LADES@MNP catalyst

Synthesis of Fe₃O₄ nanoparticles: The magnetic nanoparticle Fe₃O₄ was synthesized by co-precipitation method reported in the literature. The mixture of FeCl₂ and FeCl₃ was dissolved in 200 mL water under ultrasonic irradiation. The pH value of the solution was adjusted to 12.0 by adding amount of potassium hydroxide solution (1.0 M). After sonication for 120 mins, Fe₃O₄ was washed repeatedly water and ethanol, separated with a magnetic and drying at 80 °C for 12 hours.

Coating Fe₃O₄ nanoparticles of silica: The coating of a layer of silica on the surface of the nano Fe₃O₄ was carried out by refluxing the mixture of Fe₃O₄ (1 g), 3 mL ammonia solution and 3 mL of tetraethylorthosilicate (TEOS) in 100 mL ethanol. After 8 h, the black precipitate (SiO₂@Fe₃O₄) was collected using a magnet, followed by washing with ethanol three times and drying at 80 °C for 6 h. 5 g SiO₂@Fe₃O₄ were dispersed in 100 mL toluene by sonication for 30 min and (3-chloropropyl)triethoxysilane (4.820 g, 20.0 mmol) was then added. After refluxing for 12 hours, the reaction mixture was cooled to room temperature. The product (Fe₃O₄@SiO₂@(CH₂)₃Cl) was separated, wash with ethanol and dried for further application.

Synthesis of LADES: The reaction of urea (20.0 mol) and zinc chloride (5.0 mmol) was involved at 100 °C and stirring to afford clear solution. DES [Urea]₄[ZnCl₂] was used without further purification.

Procedure for the synthesis of LADES@MNP: The mixture of 3.0 g Fe₃O₄@SiO₂@(CH₂)₃Cl and DES [Urea]₄[ZnCl₂] (1.880 g, 5.0 mmol) were added round-bottomed flask and stirred at 90 °C for 18 hours. Lastly, the Fe₃O₄@SiO₂@(CH₂)₃-Urea-ZnCl₂ catalyst was rinsed three times with ethanol and water, collected magnetically from solution mixture, and dried under reduced pressure at 60 °C for 6 h.

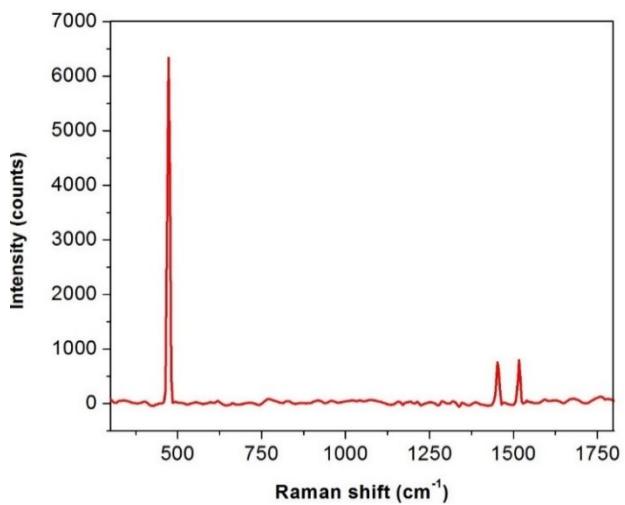


Figure S1. Raman spectra of LADES@MNP

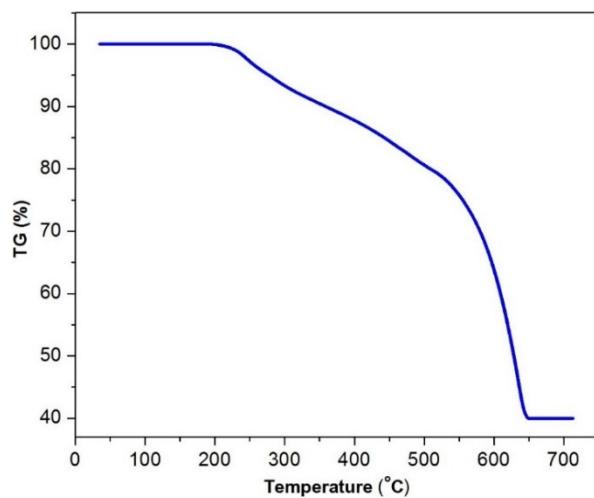


Figure S2. TG analysis of LADES@MNP

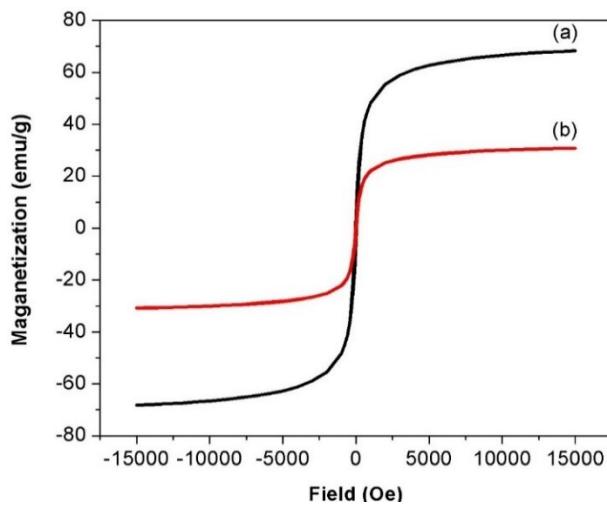
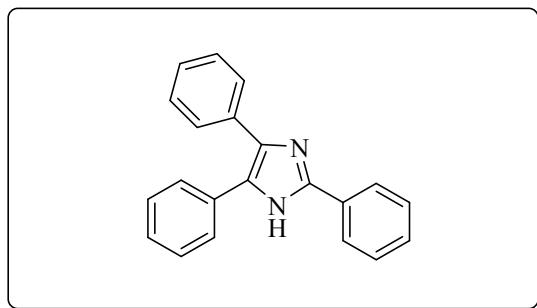


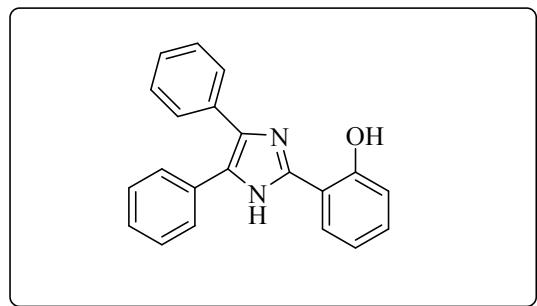
Figure S3. Magnetic curves of Fe_3O_4 (a) and LADES@MNP (b)

Section S3. Spectral data of products

2,4,5-Triphenyl-1*H*-imidazole²⁴ ($C_{21}H_{16}N_2$, **1a):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield : 123 mg (83%); mp. 276-277 °C (lit. mp. 267-269 °C) ; FT-IR (KBr) 3417, 3037, 2967, 1600, 1502, 1487, 1460, 766, 696 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.69 (s, 1H), 8.09 (d, *J* = 7.5 Hz, 2H), 7.55 (d, *J* = 7.5 Hz, 2H), 7.52-7.43 (m, 6H), 7.38 (dd, *J* = 7.0 Hz, 2H), 7.31 (dd, *J* = 7.5 Hz, 2H), 7.22 (dd, *J* = 7.0 Hz, 2H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 145.5, 137.1, 135.2, 131.1, 130.3, 128.7, 128.7, 128.5, 128.2, 128.2, 127.8, 127.1, 126.5, 125.2.

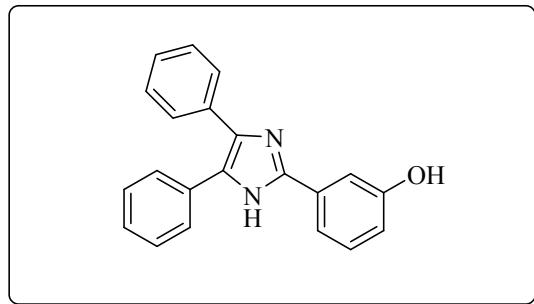


2-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenol²⁴ ($C_{21}H_{16}N_2O$, **1b):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 140 mg (90%); mp. 212-213 °C (lit. mp. 202-205 °C); FT-IR (KBr) 3421, 3213, 3061, 1607, 1494, 1443, 1383, 1261, 758, 694 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 13.01 (s, 1H), 12.93 (s, 1H), 8.01 (d, *J* = 10 Hz, 1H), 7.52-7.40 (m, 7H), 7.32 (dd, *J* = 7.5 Hz, 2H), 7.26-7.23 (m, 2H), 6.96-6.90 (m, 2H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 157.0, 146.2, 134.5, 133.9, 130.6, 130.4, 129.1, 128.9, 128.7, 127.6, 127.4, 127.1, 125.3, 119.2, 117.2, 113.2.

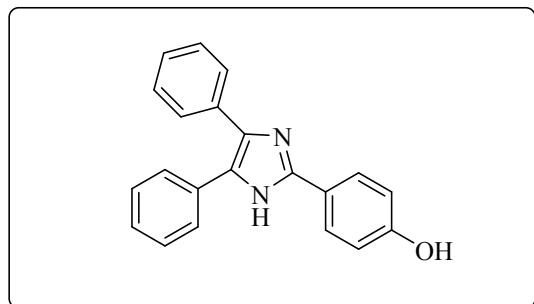


3-(4,5-Diphenyl-1*H*-imidazol-2-yl)phenol³³ ($C_{21}H_{16}N_2O$, **1c):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 125 mg (80%); mp. 263-264 °C (lit. mp. 258-260 °C) ;

FT-IR (KBr) 3372, 3060, 3017, 2914, 1618, 1502, 1230, 1193, 762, 695 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.61 (s, 1H), 9.55 (s, 1H), 7.54-7.49 (m, 5H), 7.44 (dd, *J* = 7.0 Hz, 2H), 7.37 (dd, *J* = 7.5 Hz, 2H), 7.32-7.20 (m, 4H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 157.6, 145.6, 137.0, 135.2, 131.6, 131.1, 129.7, 128.6, 128.5, 128.2, 128.1, 127.7, 127.1, 126.5, 116.1, 115.4, 112.2.

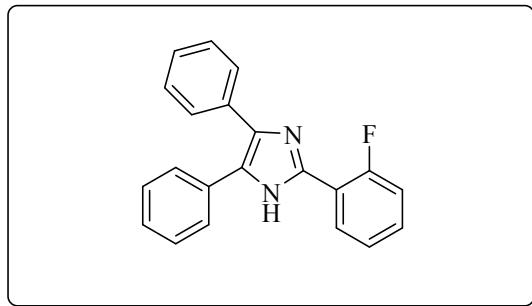


4-(4,5-Diphenyl-1H-imidazol-2-yl)phenol²⁴ (C₂₁H₁₆N₂O, 1d**):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; yellow solid; yield: 142 mg (91%); mp. 264-265 °C (lit. mp. 232-233 °C); FT-IR (KBr) 3448, 2952, 2828, 1597, 1474, 1441, 1394, 1317, 1129, 1038, 967, 764, 692 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.41 (s, 1H), 9.70 (s, 1H), 7.90 (d, *J* = 9.0 Hz, 2H), 7.54 (d, *J* = 7.5 Hz, 2H), 7.49 (d, *J* = 7.5 Hz, 2H), 7.44 (d, *J* = 7.5 Hz, 2H), 7.36 (dd, *J* = 7.0 Hz, 1H), 7.30 (dd, *J* = 7.0 Hz, 2H), 7.21(dd, *J* = 7.0 Hz, 1H), 6.85 (d, *J* = 8.5 Hz, 2H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 170.0, 157.4, 145.7, 136.3, 135.1, 131.0, 128.3, 128.0, 127.8, 127.2, 127.0, 126.7, 126.5, 126.0, 121.3, 115.1.

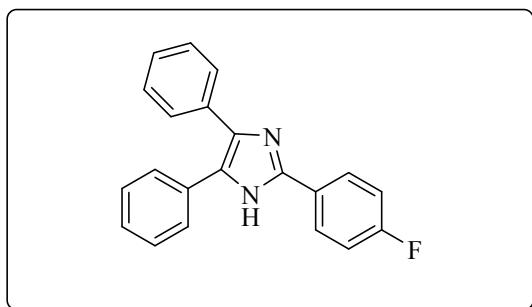


2-(2-Fluorophenyl)-4,5-diphenyl-1H-imidazole (C₂₁H₁₅FN₂, 1e**):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; yellow solid; yield: 119 mg (76%); mp. 217-218 °C; FT-IR (KBr) 3443, 3059, 2952, 2826, 1725, 1641, 1485, 1442, 1382, 1245, 1130, 1041, 766, 699 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.64 (s, 1H), 7.80 (dd, *J* = 4.0 Hz, 1H), 7.61 (dd, *J* = 4.0 Hz, 1H),

7.55 (d, $J = 7.5$ Hz, 2H), 7.50-7.46 (m, 4H), 7.43 (dd, $J = 7.0$ Hz, 2H), 7.36 (dd, $J = 7.0$ Hz, 2H), 7.31 (dd, $J = 7.5$ Hz, 2H), 7.23 (dd, $J = 7.0$ Hz, 1H) ppm; ^{13}C NMR (125 MHz, DMSO- d_6) δ 143.7, 136.3 (d, $J = 222.5$ Hz), 131.9 (d, $J = 6.3$ Hz), 131.2, 130.6 (d, $J = 4.0$ Hz), 130.4, 129.0, 128.5, 128.4, 128.1, 127.5 (d, $J = 5.0$ Hz), 126.9.

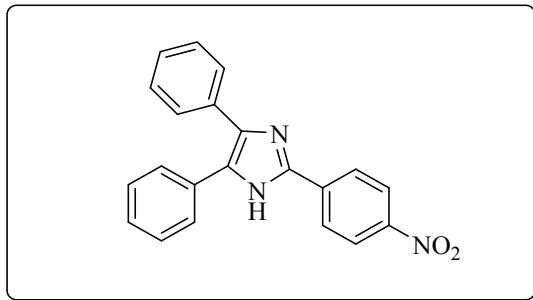


4-(4-Fluorophenyl)-4,5-diphenyl-1*H*-imidazole³⁵ ($\text{C}_{21}\text{H}_{15}\text{FN}_2$, **1f)** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 100 mg (64%); mp. 253-254 °C; FT-IR (KBr) 3418, 3030, 2856, 1606, 1541, 1494, 1444, 1382, 1322, 1226, 1023, 832, 767, 694 cm⁻¹; ^1H NMR (500 MHz, ^{13}C NMR (125 MHz, DMSO- d_6) δ) δ 12.69 (s, 1H), 8.12 (dd, $J_1 = 5.5$ Hz, $J_2 = 7.5$ Hz, 2H), 7.54 (d, $J = 7.5$ Hz, 2H), 7.50 (d, $J = 7.0$ Hz, 2H), 7.45 (dd, $J = 7.5$ Hz, 2H), 7.39-7.29 (m, 5H), 7.22 (dd, $J = 7.5$ Hz, 1H); ^{13}C NMR (125 MHz, DMSO- d_6) δ 162.1 (d, $J = 245.0$ Hz), 144.7, 137.1, 135.1, 131.0, 128.7, 128.4, 128.2 (d, $J = 30.0$ Hz), 127.8, 127.4 (d, $J = 7.5$ Hz), 127.1, 126.6, 115.7 (d, $J = 21.0$ Hz).

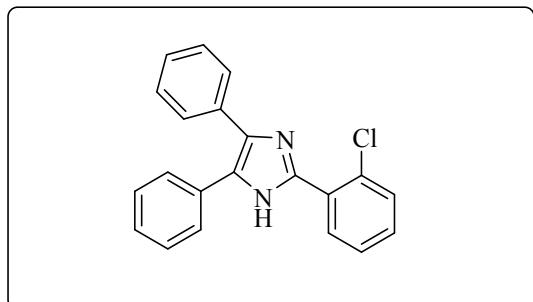


2-(4-Nitrophenyl)-4,5-diphenyl-1*H*-imidazole²⁴ ($\text{C}_{21}\text{H}_{15}\text{N}_3\text{O}_2$, **1g):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; orange solid; yield: 140 mg (82%); mp. 202-203 °C (lit. mp. 199-201 °C); FT-IR (KBr) 3418, 3030, 2856, 1606, 1541, 1494, 1444, 1382, 1322, 1226, 1023, 832, 768, 694 cm⁻¹; ^1H NMR (500 MHz, DMSO- d_6) δ 13.50 (s, 1H), 8.40-8.39 (m, 5H), 8.29-8.24 (m, 4H), 7.86-7.81 (m, 3H), 7.72 (d, $J = 4$ Hz, 1H), 7.51-7.46 (m, 2H); ^{13}C NMR (125 MHz, DMSO-

d_6) δ 159.4, 136.9, 132.8, 131.8, 131.2, 130.9, 129.3, 129.0, 128.8, 128.5, 128.2, 127.7, 127.4, 126.5, 126.2, 126.1, 124.1, 122.7.

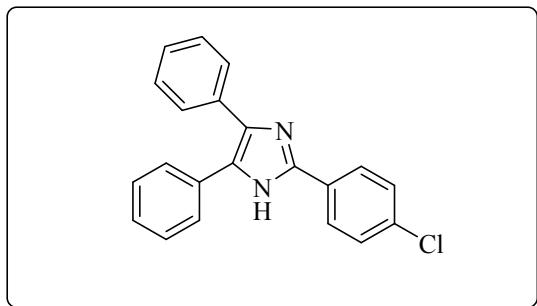


2-(2-Chlorophenyl)-4,5-diphenyl-1*H*-imidazole³² ($C_{21}H_{15}ClN_2$, **1h):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 145 mg (88%); mp. 212-214 °C (lit. mp. 190-191 °C); FT-IR (KBr) 3415, 2951, 2826, 1601, 1440, 1392, 1315, 1196, 1128, 1038, 966, 907, 763, 691 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.64 (s, 1H), 7.81-7.80 (m, 1H), 7.62-7.60 (m, 1H); 7.55-7.42 (m, 8H), 7.38-7.29 (m, 3H), 7.23 (dd, *J* = 7.0 Hz, 1H) ppm; ¹³C NMR (125 MHz, DMSO-*d*₆) δ 143.1, 136.5, 134.8, 131.3, 131.2, 130.6, 129.9, 129.8, 129.7, 128.3, 127.9, 127.4, 126.9, 126.3.

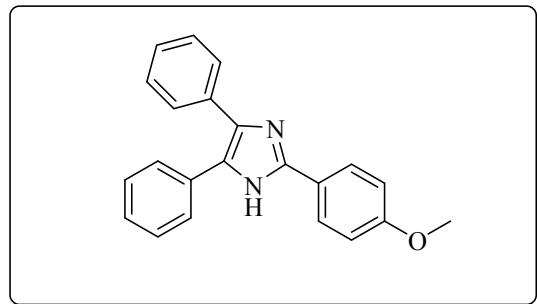


2-(4-Chlorophenyl)-4,5-diphenyl-1*H*-imidazole²⁴ ($C_{21}H_{15}ClN_2$, **1i):** Analytical TLC on silica gel, 1:9ethyl acetate/hexane; white solid; yield: 141 mg (85%); mp. 267-268 °C (lit. mp. 262-264 °C) ; FT-IR (KBr) 3415, 1603, 1485, 1433, 1128, 1092, 830, 765, 696 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.78 (s, 1H), 8.09 (d, *J* = 8.0 Hz, 2H), 7.54-7.43 (m, 7H), 7.39 (d, *J* = 7.0 Hz, 1H), 7.31 (dd, *J* = 7.0 Hz, 2H), 7.23 (d, *J* = 7.0 Hz, 1H) ppm; ¹³C NMR (125 MHz, DMSO-*d*₆) δ

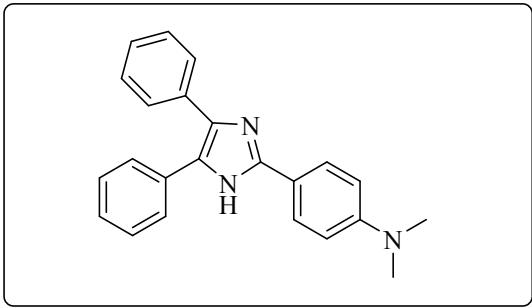
144.7, 137.6, 135.3, 133.1, 131.2, 129.5, 129.1, 129.0, 128.9, 128.8, 128.5, 128.2, 127.4, 127.2, 127.0.



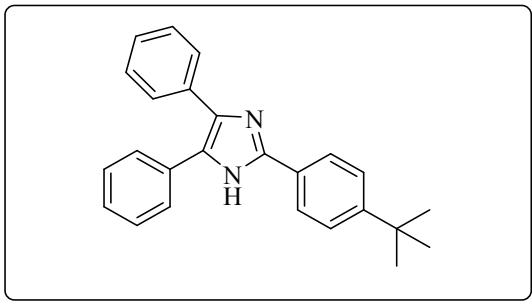
2-(4-Methoxyphenyl)-4,5-diphenyl-1*H*-imidazole²⁴ ($C_{22}H_{18}N_2O$, **1k):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 137 mg (84%); mp. 258-259 °C (lit. mp. 220-223 °C); FT-IR (KBr) 3416, 2959, 2837, 1612, 1492, 1443, 1249, 1200, 1030, 968, 829, 765, 695 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.51 (s, 1H), 8.01 (d, *J* = 8.0 Hz, 2H), 7.54 (d, *J* = 7.5 Hz, 2H), 7.49 (d, *J* = 7.5 Hz, 2H), 7.43 (dd, *J* = 7.0 Hz, 2H), 7.36 (dd, *J* = 7.5 Hz, 1H), 7.30 (dd, *J* = 7.5 Hz, 2H), 7.21 (dd, *J* = 7.5 Hz, 1H), 7.04 (d, *J* = 7.5 Hz, 2H), 3.81 (s, 3H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 159.4, 145.6, 136.8, 135.3, 131.2, 128.6, 128.4, 128.2, 127.6, 127.6, 127.0, 126.7, 126.4, 123.1, 114.1, 55.2.



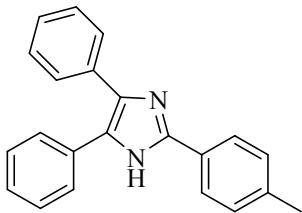
4-(4,5-Diphenyl-1*H*-imidazol-2-yl)-*N,N*-dimethylaniline³³ ($C_{23}H_{21}N_3$, **1l):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 149 mg (88%); mp. 258-259 °C (lit. mp. 256-259 °C); FT-IR (KBr) 3417, 2797, 1616, 1496, 1444, 819, 765, 696 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.31 (s, 1H), 7.90 (d, *J* = 9.0 Hz, 2H), 7.53 (d, *J* = 7.0 Hz, 2H), 7.48 (d, *J* = 7.0 Hz, 2H), 7.42 (t, *J* = 7.0 Hz, 2H), 7.35 (d, *J* = 7.0 Hz, 1H), 7.29 (t, *J* = 7.0 Hz, 2H), 7.21 (d, *J* = 7.0 Hz, 1H), 6.79 (d, *J* = 9.0 Hz, 2H), 2.96 (s, 6H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 150.3, 146.5, 136.5, 135.5, 131.4, 128.6, 128.3, 128.1, 127.4, 127.0, 126.3, 118.3, 111.9, 39.9.



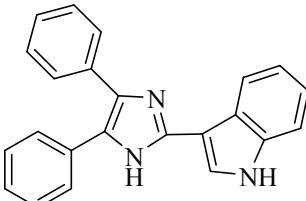
2-(4-(*tert*-Butyl)phenyl)-4,5-diphenyl-1*H*-imidazole ($C_{24}H_{22}N_2$, **1m):** Analytical TLC on silica gel, 1:9 ethyl acetate/*n*-hexane; white solid; yield: 165 mg (94%); mp. 285-286 °C; FT-IR (KBr) 3416, 3057, 2963, 2867, 1603, 1492, 1450, 1385, 1268, 976, 838, 765, 696 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.61 (s, 1H), 8.01 (d, *J* = 8.0 Hz, 2H), 7.55 (d, *J* = 7.0 Hz, 2H), 7.50 (d, *J* = 6.0 Hz, 4H), 7.44 (d, *J* = 7.0 Hz, 2H), 7.38 (d, *J* = 7.0 Hz, 1H), 7.30 (dd, *J* = 7.0 Hz, 2H), 7.23 (d, *J* = 7.0 Hz, 1H), 1.32 (s, 9H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 151.1, 145.9, 137.3, 135.6, 131.5, 129.0, 128.8, 128.5, 128.3, 128.0, 127.4, 126.8, 125.7, 125.3, 34.76, 31.4.



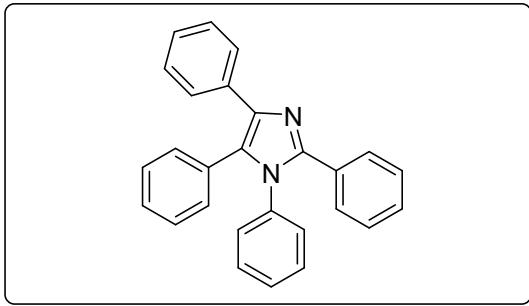
4,5-Diphenyl-2-(*p*-tolyl)-1*H*-imidazole²⁴ ($C_{22}H_{18}N_2$, **1n):** Analytical TLC on silica gel, 1:9ethyl acetate/hexane; white solid; yield: 147 mg (95%); mp. 231-232 °C (lit. mp. 233-235 °C); FT-IR (KBr) 3416, 3028, 1602, 1493, 1449, 1384, 1322, 1200, 1127, 968, 822, 765, 695 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.59 (s, 1H), 7.97 (d, *J* = 8.0 Hz, 2H), 7.55 (d, *J* = 7.5 Hz, 2H), 7.51 (d, *J* = 7.5 Hz, 2H), 7.44 (dd, *J* = 7.5 Hz, 2H), 7.37 (dd, *J* = 7.5 Hz, 1H), 7.31-7.28 (m, 4H), 7.22 (dd, *J* = 7.5 Hz, 1H), 2.35 (s, 3H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 146.0, 138.0, 137.2, 135.6, 131.5, 129.6, 129.0, 128.7, 128.5, 128.3, 128.0, 127.4, 126.8, 125.5, 21.2.



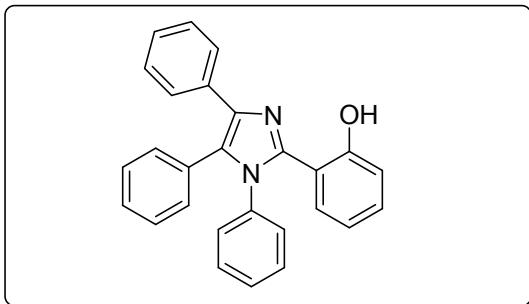
3-(4,5-Diphenyl-1*H*-imidazol-2-yl)-1*H*-indole ($C_{23}H_{17}N_3$, **1o):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 156 mg (93%); mp. 306-307 °C; FT-IR (KBr) 3413, 3049, 2959, 1606, 1492, 1443, 1335, 1245, 846, 757, 694 cm^{-1} ; ^1H NMR (500 MHz, DMSO- d_6) δ 12.27 (s, 1H), 11.37 (s, 1H), 8.48 (d, $J = 7.5$ Hz, 1H), 8.00 (s, 1H), 7.64 (d, $J = 7.5$ Hz, 2H), 7.52 (d, $J = 7.0$ Hz, 2H), 7.46-7.43 (m, 3H), 7.37-7.31 (m, 3H), 7.23-7.14 (m, 3H); ^{13}C NMR (125 MHz, DMSO- d_6) δ 143.4, 135.9, 135.6, 135.4, 131.3, 128.4, 127.8, 127.1, 126.6, 125.8, 125.4, 124.8, 123.4, 121.5, 121.2, 119.4, 111.3, 106.5.



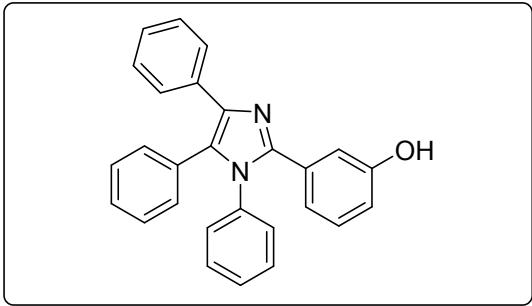
1,2,4,5-Tetraphenyl-1*H*-imidazole³² ($C_{27}H_{20}N_2$, **2a):** Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 136 mg (73%), mp. 213-217 °C; FT-IR (KBr, 4000-400 cm^{-1}): 3419, 3054, 1594, 1490, 1446, 1320 cm^{-1} ; ^1H NMR (500 MHz, CDCl₃- d_1) δ 7.43-7.42 (m, 8H), 7.42-7.04 (m, 11H); ^{13}C NMR (125 MHz, CDCl₃- d_1) δ 147.1, 137.3, 134.6, 131.3, 131.0, 130.9, 130.7, 129.2, 129.1, 128.6, 128.5, 128.4, 128.3, 128.2, 128.1, 128.0, 127.6, 126.7, 125.4.



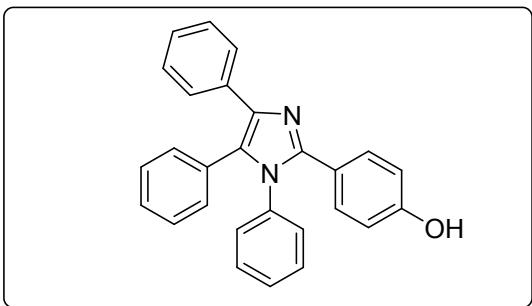
2-(2-Hydroxyphenyl)-1,4,5-triphenyl imidazole¹⁷ (C₂₇H₂₀N₂O, 2b): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 151 mg (78%), mp. 250-254 °C; FT-IR (KBr) 3420, 3060, 2930, 1645, 1591, 1487, 1381, 1252 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 12.57 (s, 1H), 7.45-7.29 (m, 16H), 6.94 (d, *J* = 8.0 Hz, 1H), 6.67-6.62 (m, 1H), 6.55 (t, *J* = 8.0 Hz, 1H); ¹³C NMR (125 MHz, DMSO-d₆) δ 157.3, 144.4, 136.6, 134.4, 133.2, 131.3, 130.8, 130.2, 129.6, 129.4, 129.3, 128.7, 128.7, 128.5, 128.4, 126.9, 126.8, 126.1, 118.1, 116.9, 113.9.



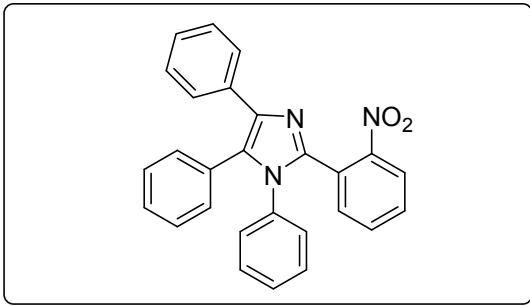
2-(3-Hydroxyphenyl)-1,4,5-triphenyl imidazole³⁴ (C₂₇H₂₀N₂O, 2c): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 155 mg (80%), mp. 202-205 °C; FT-IR (KBr) 3400, 3057, 1594, 1490, 1448, 1396, 1320 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 9.50 (s, 1H), 7.48 (d, *J* = 7.7 Hz, 2H), 7.32-7.15 (m, 13H), 7.17 (t, *J* = 7.1 Hz, 1H), 6.68 (t, *J* = 8.7 Hz, 2H); ¹³C NMR (125 MHz, DMSO-d₆) δ 157.1, 146.1, 136.7, 136.7, 134.4, 131.5, 131.2, 131.2, 130.4, 129.1, 129.1, 128.7, 128.4, 128.4, 128.2, 126.4, 126.3, 118.9, 115.4, 115.4.



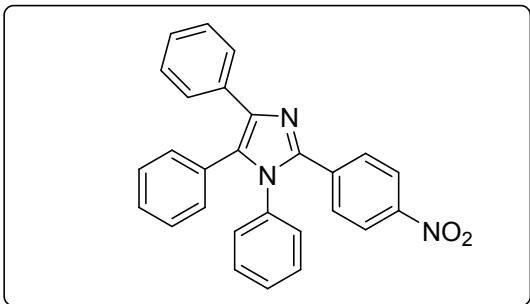
2-(4-Hydroxyphenyl)-1,4,5-triphenyl imidazole³² (C₂₇H₂₀N₂O, 2d): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 146 mg (75%), mp. 285-289 °C; FT-IR (KBr) 3423, 2923, 1598, 1456, 1379, 1270 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 9.67 (s, 1H), 7.47-7.22 (m, 17H), 6.65 (s, 2H); ¹³C NMR (125 MHz, DMSO-d₆) δ 157.6, 146.4, 136.9, 136.4, 134.6, 131.1, 130.6, 130.6, 129.8, 129.1, 128.8, 128.6, 128.4, 128.3, 128.1, 126.3, 121.3, 114.9.



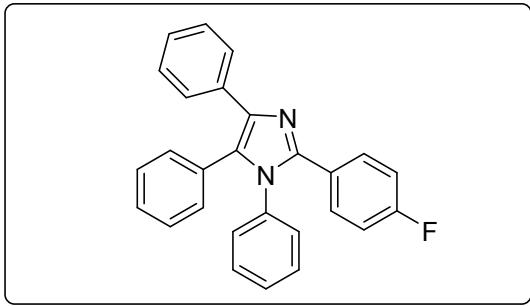
2-(2-Nitrophenyl)-1,4,5-triphenyl imidazole³³ (C₂₇H₁₉N₃O₂, 2e): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 146 mg (70%); mp. 213-216 °C; FT-IR (KBr) 3427, 3060, 1593, 1509, 1398, 1343 cm⁻¹; ¹H NMR (500 MHz, CDCl₃-d₁) δ 7.90 (d, *J* = 8.0 Hz, 1H), 7.67 (d, *J* = 7.5 Hz, 1H), 7.60 (dd, *J* = 19.9, 7.5 Hz, 3H), 7.50 (t, *J* = 8.0 Hz, 1H), 7.26-7.14 (m, 11H), 6.94 (d, *J* = 7.5 Hz, 2H); ¹³C NMR (125 MHz, CDCl₃-d₁) δ 149.3, 143.7, 139.1, 136.1, 134.6, 133.8, 133.3, 131.4, 131.0, 130.7, 130.3, 129.3, 128.8, 128.6, 128.4, 127.8, 127.1, 127.0, 124.8.



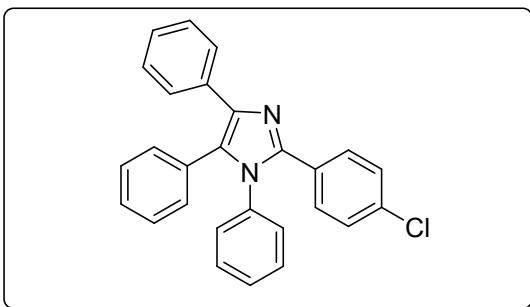
2-(4-Nitrophenyl)-1,4,5-triphenyl imidazole³³ ($C_{27}H_{19}N_3O_2$, 2f): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 188 mg (90%), mp. 194-198 °C; FT-IR (KBr) 3430, 3054, 2926, 2851, 1594, 1449, 1339, 1241 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 8.15 (d, *J* = 8.5 Hz, 2H), 7.62 (d, *J* = 8.5 Hz, 2H), 7.51 (d, *J* = 7.5 Hz, 2H), 7.39-7.27 (m, 12H), 7.21 (t, *J* = 7.5 Hz, 1H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 146.7, 143.8, 137.8, 136.3, 136.2, 133.9, 132.8, 131.1, 129.9, 129.5, 129.3, 128.8, 128.7, 128.6, 128.5, 128.3, 126.8, 126.4, 123.5.



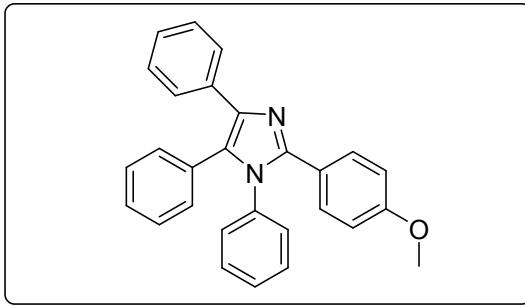
2-(4-Florophenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{19}N_2F$, 2g): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 164 mg (84%), mp. 194-197 °C; FT-IR (KBr) 3434, 3050, 1596, 1487, 1378, 1224 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 7.48 (d, *J* = 8.0 Hz, 2H), 7.42-7.39 (m, 2H), 7.34-7.23 (m, 6H), 7.27-7.23 (m, 6H), 7.19-7.13 (m, 6H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 161.9 (d, *J* = 246.25 Hz), 145.2, 136.8, 136.5, 135.6, 134.3, 131.3, 131.1, 130.5, 130.4, 130.5, 130.3, 129.5 (d, *J* = 11.13 Hz), 129.2, 128.8, 128.7, 128.5, 128.4, 128.2, 126.9 (d, *J* = 3.38 Hz), 126.5, 126.3, 115.2 (d, *J* = 21.5 Hz).



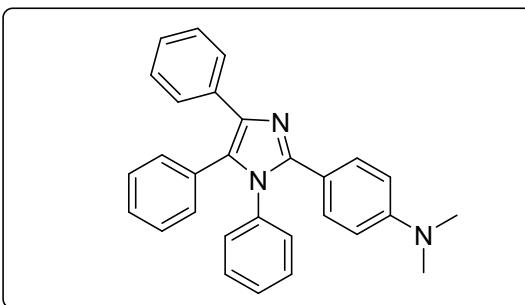
2-(4-Chlorophenyl)-1,4,5-triphenyl imidazole³² (C₂₇H₁₉N₂Cl, 2h): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 150 mg (74%), mp. 184-188 °C; FT-IR (KBr) 3057, 1595, 1487, 1404 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 7.49 (d, *J* = 8.0 Hz, 2H), 7.37-7.24 (m, 16H), 7.18 (dd, *J* = 7.5 Hz, 1H); ¹³C NMR (125 MHz, DMSO-d₆) δ 145.4, 137.5, 136.9, 134.7, 133.6, 132.1, 131.6, 130.7, 130.3, 129.7, 129.7, 129.4, 129.2, 129.0, 128.9, 128.8, 128.7, 127.0, 126.8.



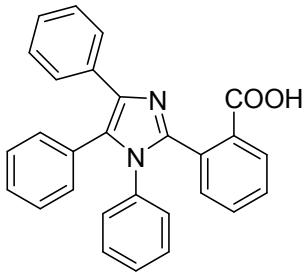
2-(4-Methoxyphenyl)-1,4,5-triphenyl imidazole³² (C₂₈H₂₂N₂O, 2k): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 143 mg (71%), mp. 174-177 °C; FT-IR (KBr) 3417, 3053, 2945, 1603, 1484, 1378, 1247 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 7.48 (d, *J* = 7.5 Hz, 2H), 7.34-7.28 (m, 8H), 7.24 (dd, *J* = 7.5 Hz, 6H), 7.16 (dd, *J* = 7.5 Hz, 1H), 6.84 (d, *J* = 8.5 Hz, 2H), 3.72 (s, 3H); ¹³C NMR (125 MHz, DMSO-d₆) δ 159.2, 146.0, 136.8, 136.5, 134.5, 131.1, 130.8, 130.5, 129.6, 129.1, 128.8, 128.7, 128.4, 128.3, 128.1, 126.3, 122.8, 113.6, 55.1.



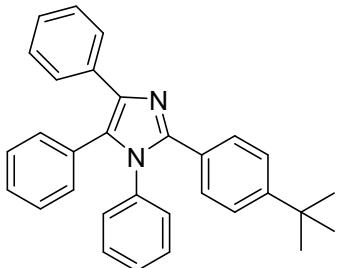
2-(4-N,N-dimethylaminophenyl)-1,4,5-triphenyl imidazole (C₂₉H₂₅N₃, 2l): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 187 mg (90%), mp. 207-209 °C; FT-IR (KBr) 3416, 2804, 1608, 1488, 1439, 1367, 1194 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 7.47 (d, *J* = 7.5 Hz, 2H), 7.33-7.27 (m, 7H), 7.23-7.17 (m, 8H), 6.57 (d, *J* = 8.5 Hz, 2H), 2.88 (s, 6H); ¹³C NMR (125 MHz, DMSO-d₆) δ 149.9, 146.7, 137.1, 136.3, 134.7, 131.2, 130.7, 130.4, 129.1, 129.0, 128.8, 128.5, 128.4, 128.2, 128.1, 126.3, 126.2, 117.7, 111.3.



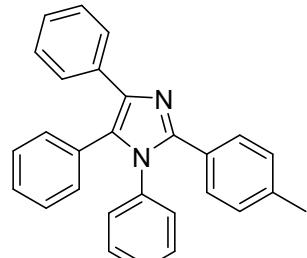
2-(2-Carboxyphenyl)-1,4,5-triphenyl imidazole (C₂₈H₂₀N₂O₂, 2m): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 193 mg (93%), mp. 294-298 °C; FT-IR (KBr) 3416, 3059, 2848, 1598, 1497, 1368, 1238 cm⁻¹; ¹H NMR (500 MHz, DMSO-d₆) δ 7.90-7.83 (m, 3H), 7.64 (t, *J* = 6.0 Hz, 1H), 7.52-7.51 (m, 7H), 7.40-7.37 (m, 5H), 7.33-7.31 (m, 3H); ¹³C NMR (125 MHz, DMSO-d₆) δ 169.0, 144.9, 132.7, 130.7, 130.2, 129.0, 128.8, 128.6, 128.5, 127.6, 127.3.



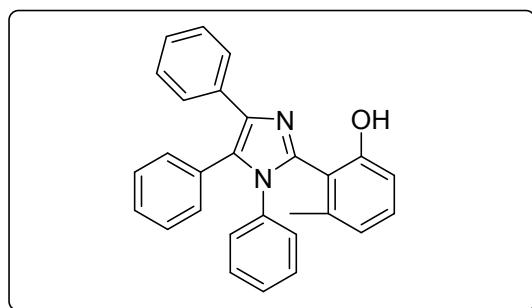
2-(4-*tert*-Butylphenyl)-1,4,5-triphenyl imidazole ($C_{31}H_{29}N_2$, 2o): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 188 mg (88%), mp. 193-196 °C; FT-IR (KBr) 3430, 3056, 2957, 1598, 1491, 1266 cm^{-1} ; ^1H NMR (500 MHz, $\text{DMSO}-d_6$) δ 7.48 (d, $J = 7.5$ Hz, 2H), 7.35-7.15 (m, 17H), 1.24 (s, 9H); ^{13}C NMR (125 MHz, $\text{DMSO}-d_6$) δ 150.8, 146.0, 136.8, 136.7, 134.5, 131.2, 131.2, 130.5, 129.2, 128.8, 128.5, 128.4, 128.2, 127.9, 127.6, 126.5, 126.4, 125.0, 34.4, 31.0.



2-(4-Methylphenyl)-1,4,5-triphenyl imidazole³³ ($C_{28}H_{22}N_2$, 2p): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 145 mg (75%), mp. 187-190 °C; FT-IR (KBr) 3427, 3047, 2920, 1593, 1489, 1442, 1377, 1320 cm^{-1} ; ^1H NMR (500 MHz, $\text{DMSO}-d_6$) δ 7.50-7.47 (m, 2H), 7.31 (m, 6H), 7.27-7.24 (m, 9H), 7.09 (d, $J = 8.0$ Hz, 2H), 2.26 (s, 3H); ^{13}C NMR (125 MHz, $\text{DMSO}-d_6$) δ 146.1, 137.8, 136.7, 136.7, 134.5, 131.1, 131.1, 130.5, 129.1, 128.8, 128.7, 128.7, 128.4, 128.4, 128.2, 128.1, 127.6, 126.4, 126.4, 20.7.



2-(2-Hydroxy-5-methylphenyl)-1,4,5-triphenyl imidazole (C₂₈H₂₃N₂O, 2q): Analytical TLC on silica gel, 1:9 ethyl acetate/hexane; white solid; yield: 187 mg (93%), mp. 200-202 °C; FT-IR (KBr) 3428, 3055, 2914, 2793, 1596, 1495, 1379, 1247 cm⁻¹; ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.32 (s, 1H), 7.44-7.20 (m, 15H), 6.98 (d, *J* = 8.0 Hz, 1H), 6.83 (d, *J* = 8.0 Hz, 1H), 6.40 (s, 1H), 1.89 (s, 3H); ¹³C NMR (125 MHz, DMSO-*d*₆) δ 155.1, 144.6, 136.7, 134.4, 133.2, 131.3, 130.7, 130.6, 129.7, 129.3, 129.1, 128.7, 128.7, 128.5, 128.4, 127.2, 126.9, 126.3, 126.1, 116.7, 113.4, 20.1.



Section S4. Copies of ^1H and ^{13}C NMR spectra of all products

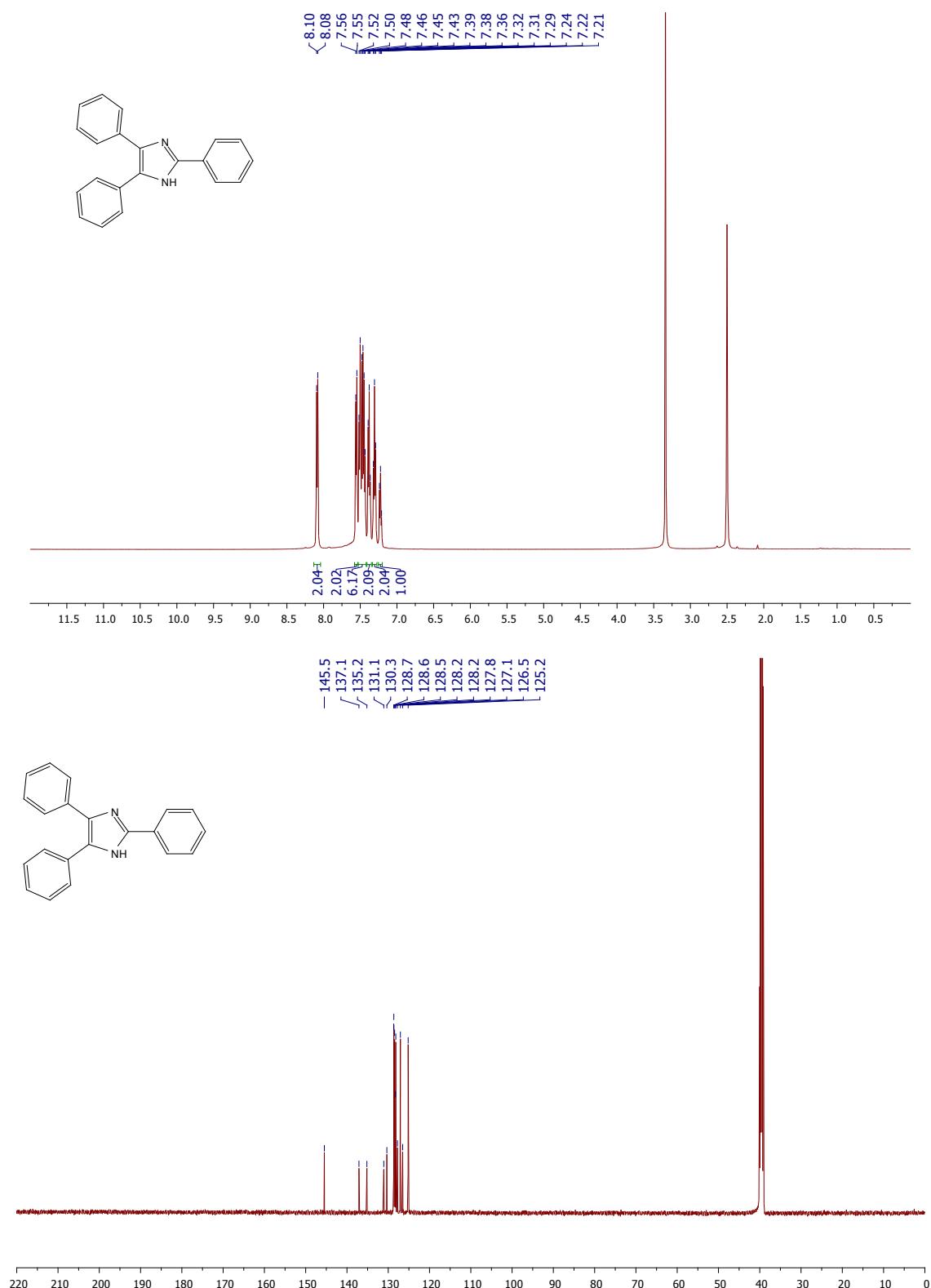


Figure S1. ^1H (top) and ^{13}C (bottom) NMR spectra of 2,4,5-triphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{16}\text{N}_2$, **1a**)

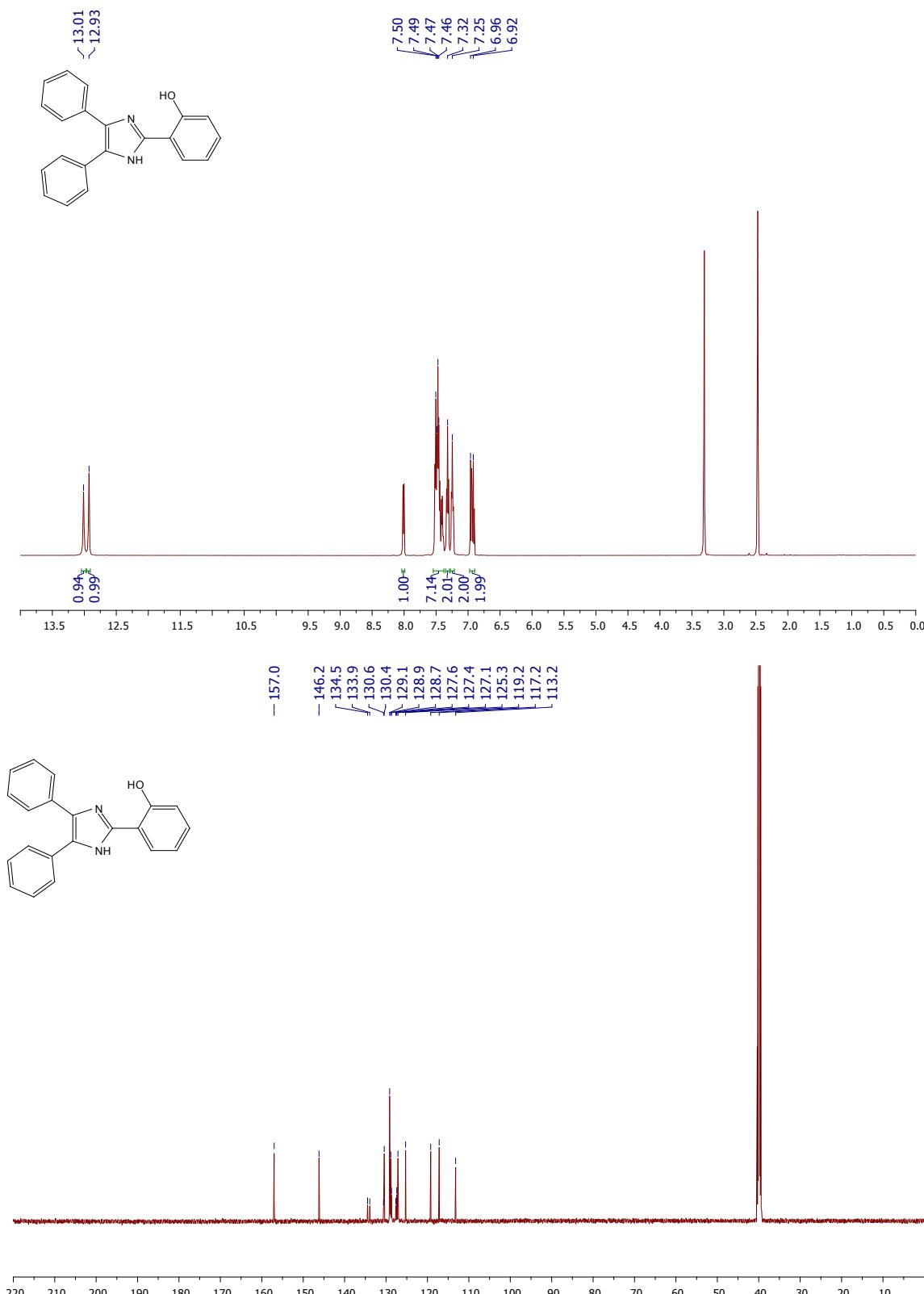


Figure S2. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol ($\text{C}_{21}\text{H}_{16}\text{N}_2\text{O}$, 1b)

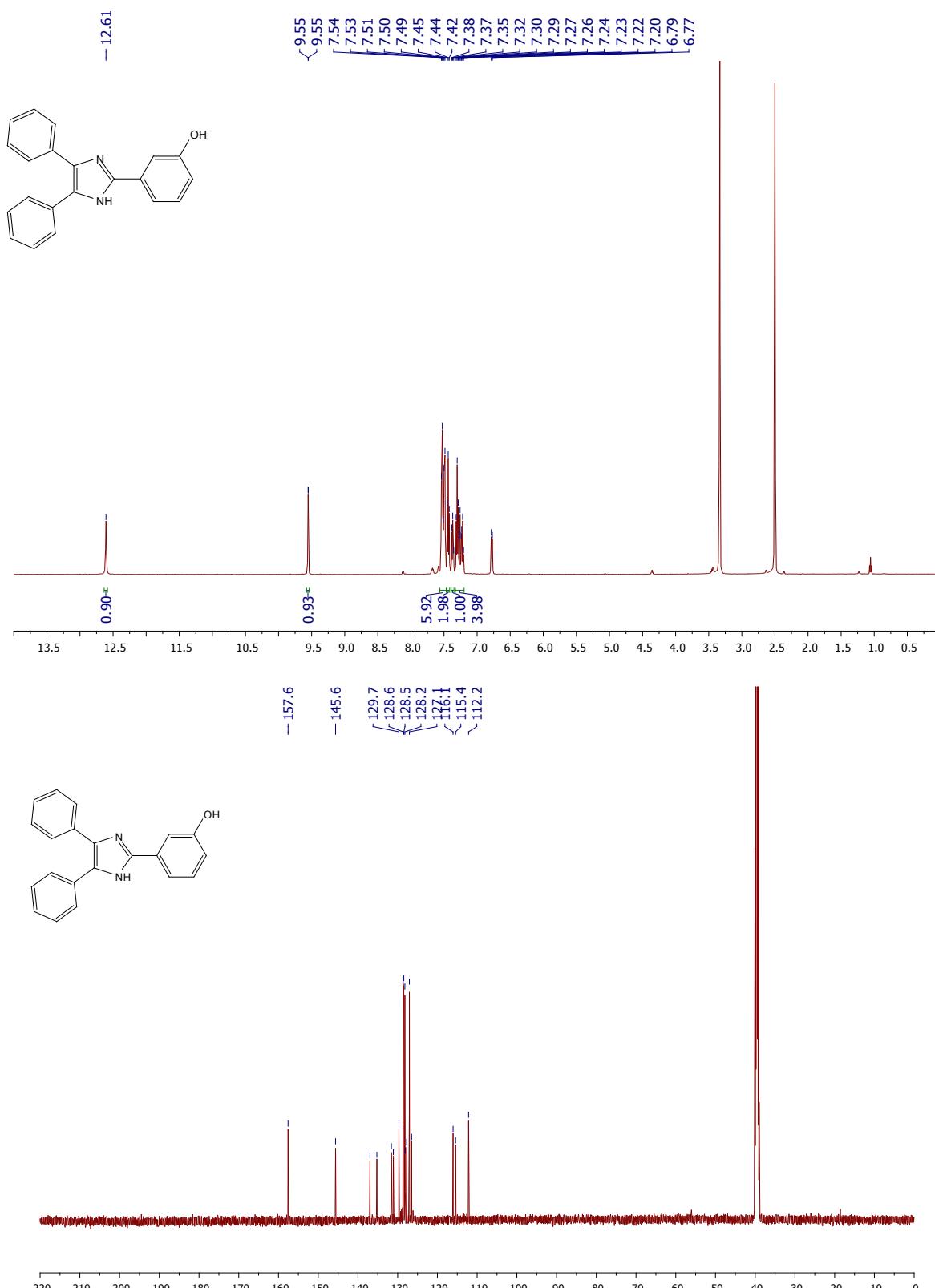


Figure S3. ^1H (top) and ^{13}C (bottom) NMR spectra of 3-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol ($\text{C}_{21}\text{H}_{16}\text{N}_2\text{O}$, 1c)

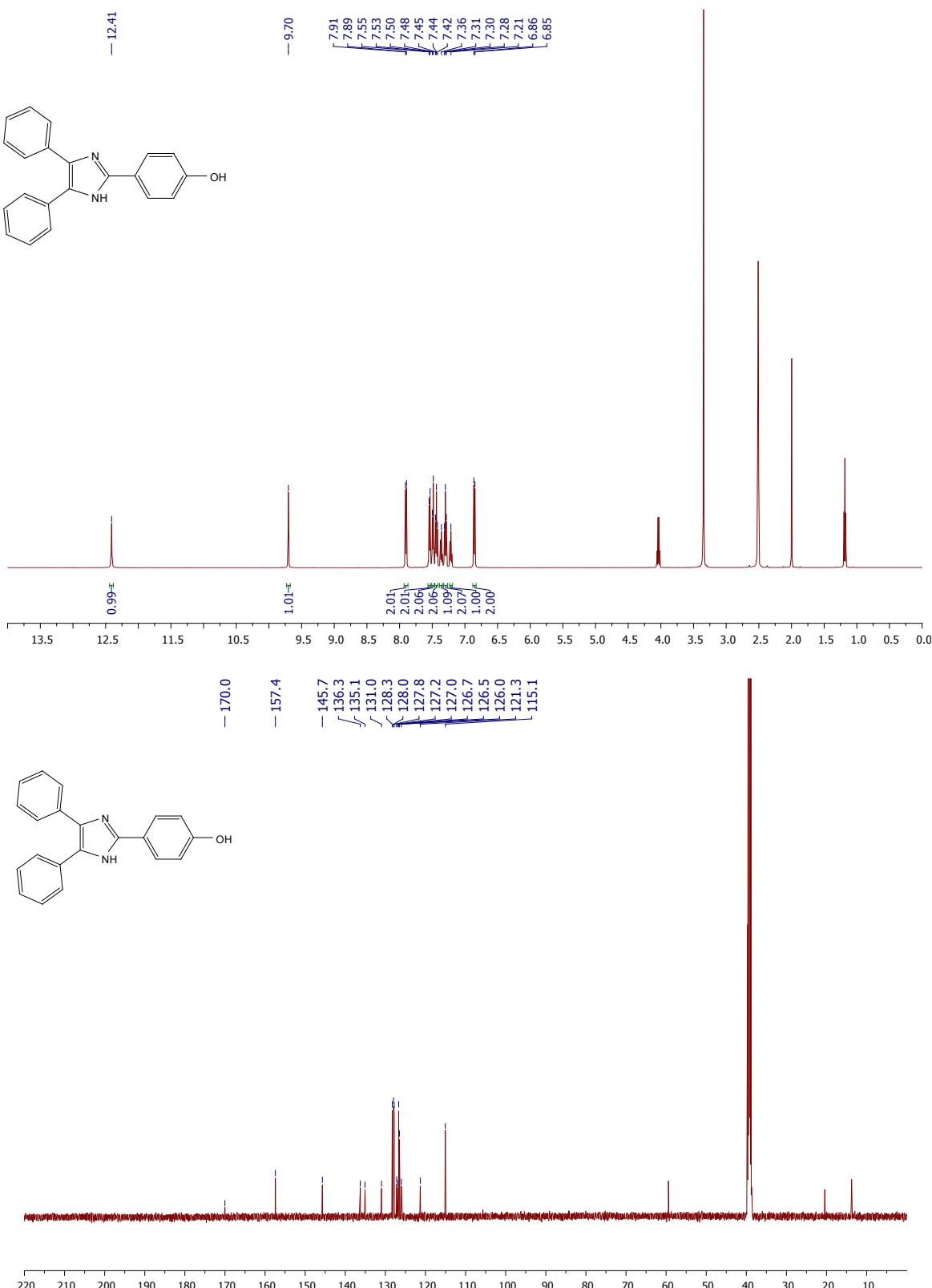


Figure S4. ^1H (top) and ^{13}C (bottom) NMR spectra of 4-(4,5-diphenyl-1*H*-imidazol-2-yl)phenol ($\text{C}_{21}\text{H}_{16}\text{N}_2\text{O}$, **1d)**

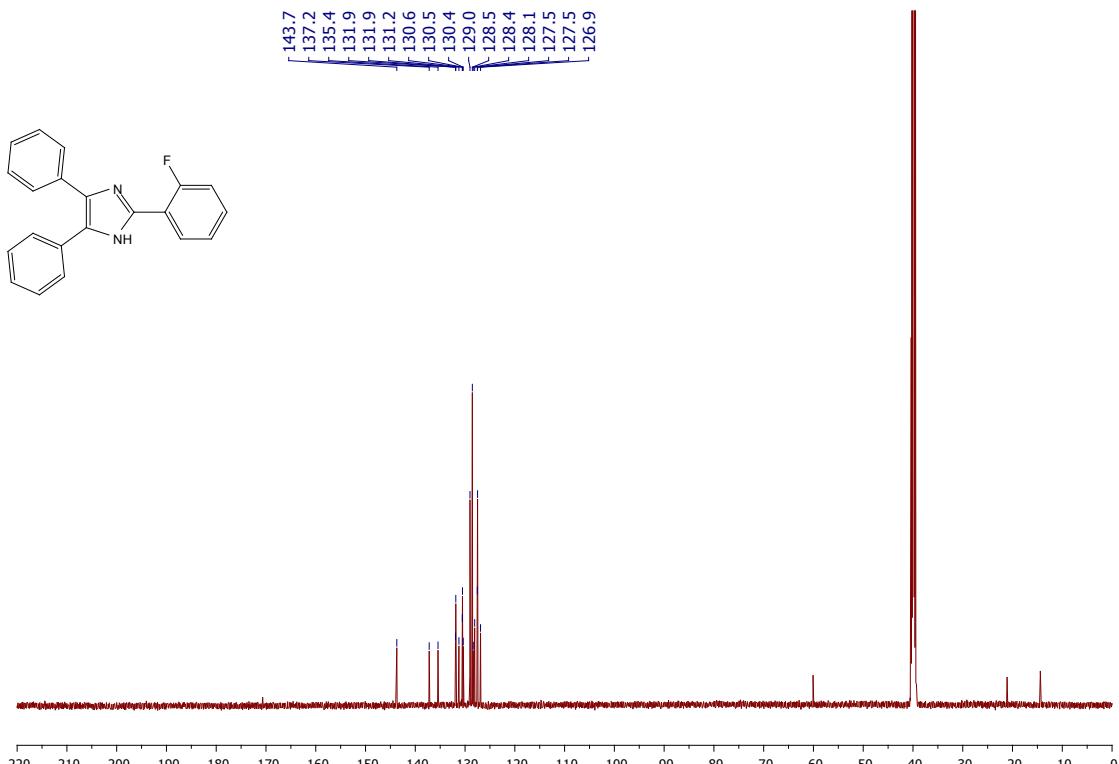
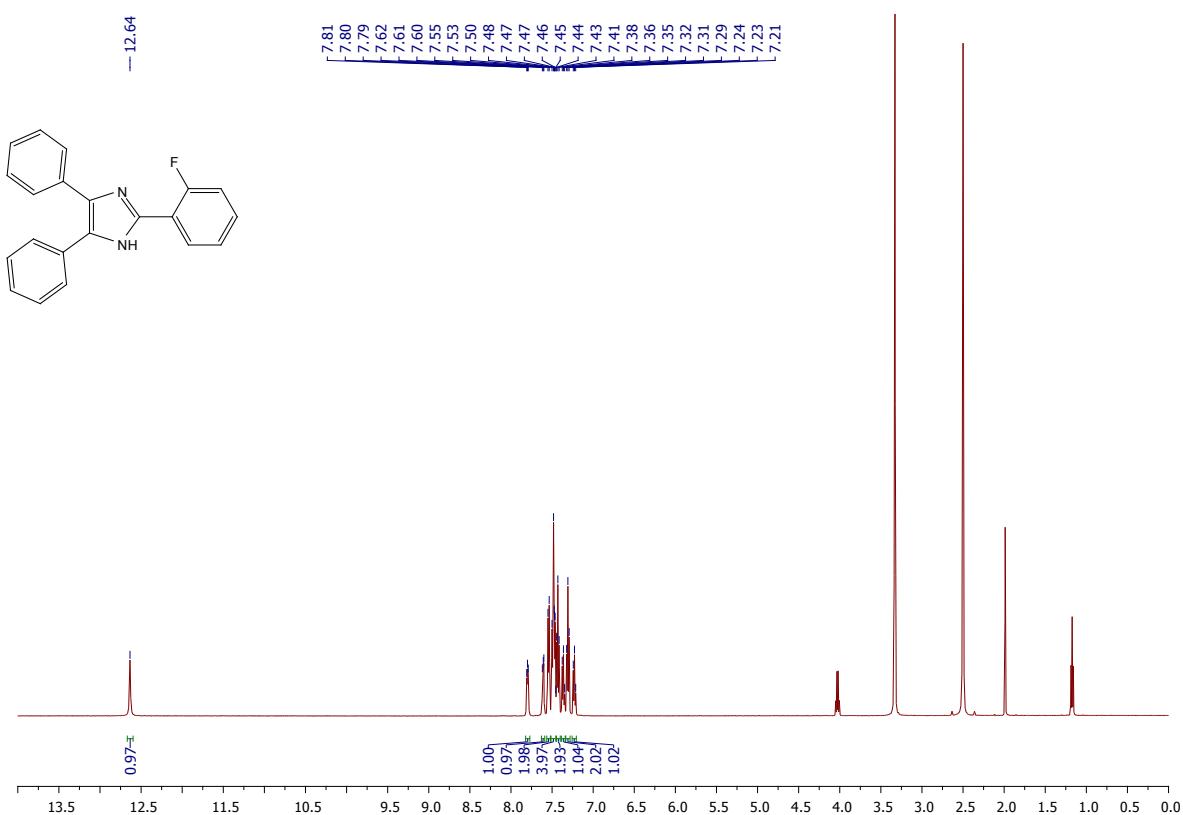


Figure S5. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(2-fluorophenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{15}\text{FN}_2$, 1e)

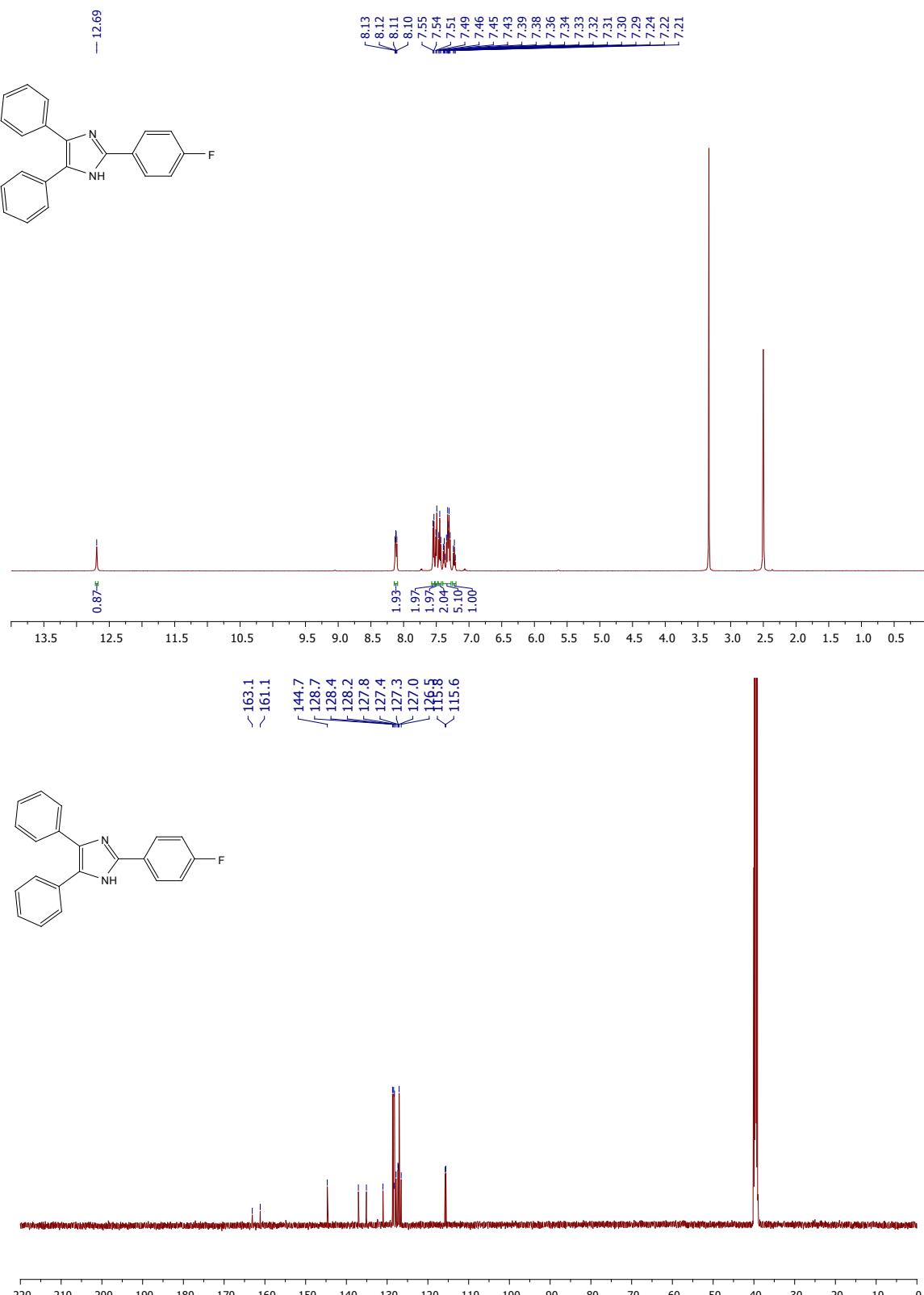


Figure S6. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-fluorophenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{15}\text{FN}_2$, 1f)

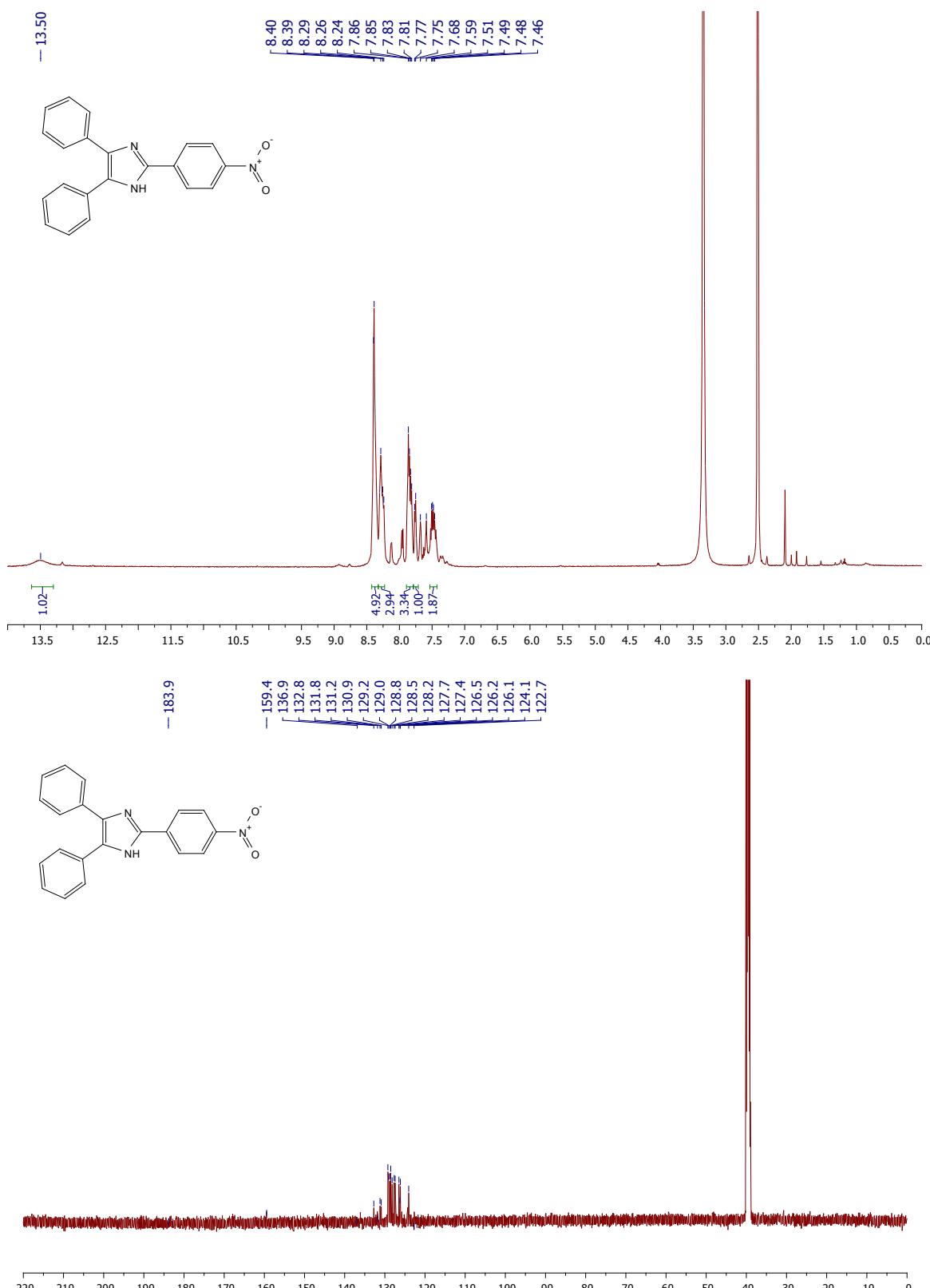


Figure S7. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-nitrophenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{15}\text{N}_3\text{O}_2$, 1g)

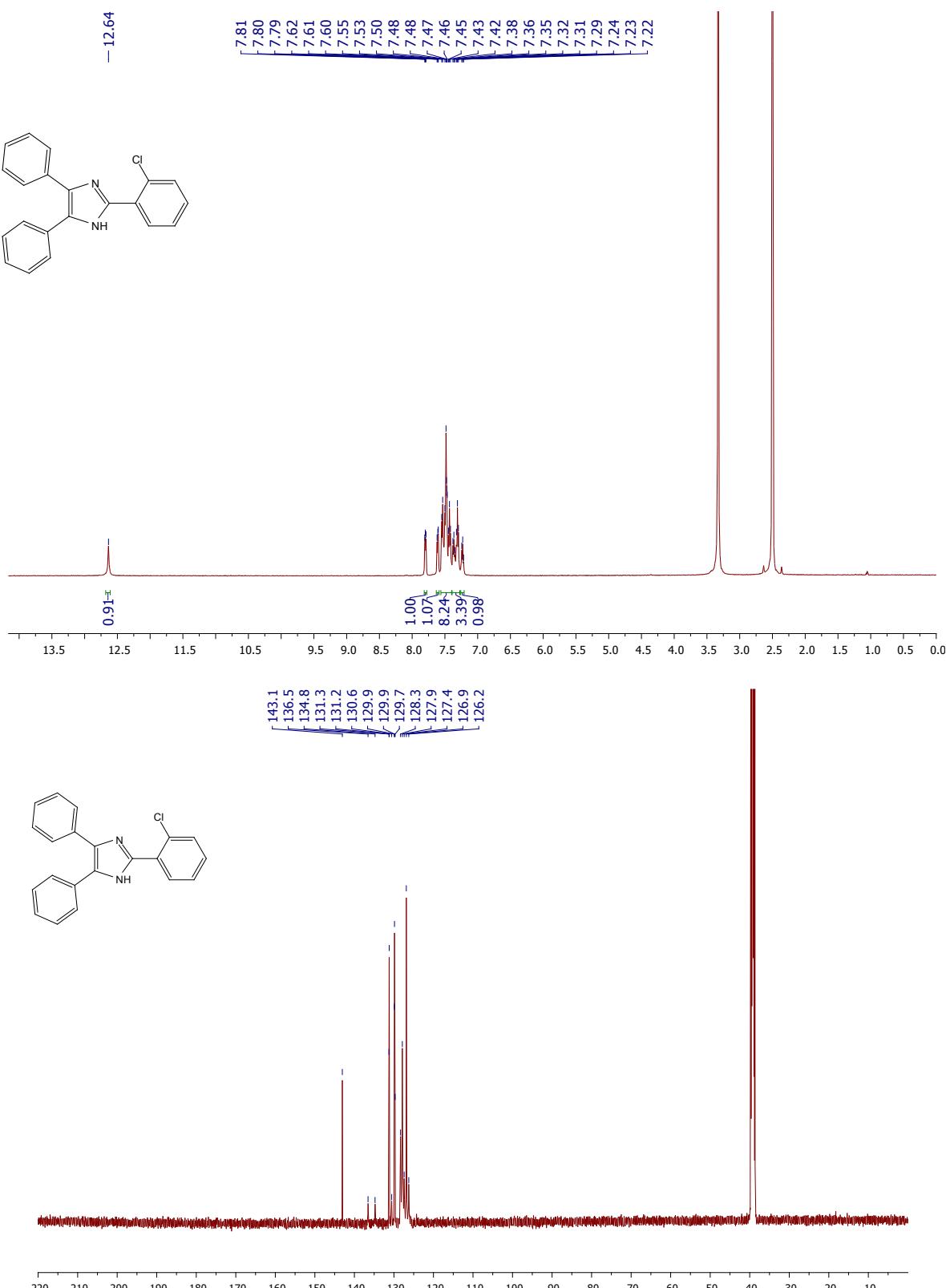


Figure S8. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(2-chlorophenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{15}\text{ClN}_2$, 1h)

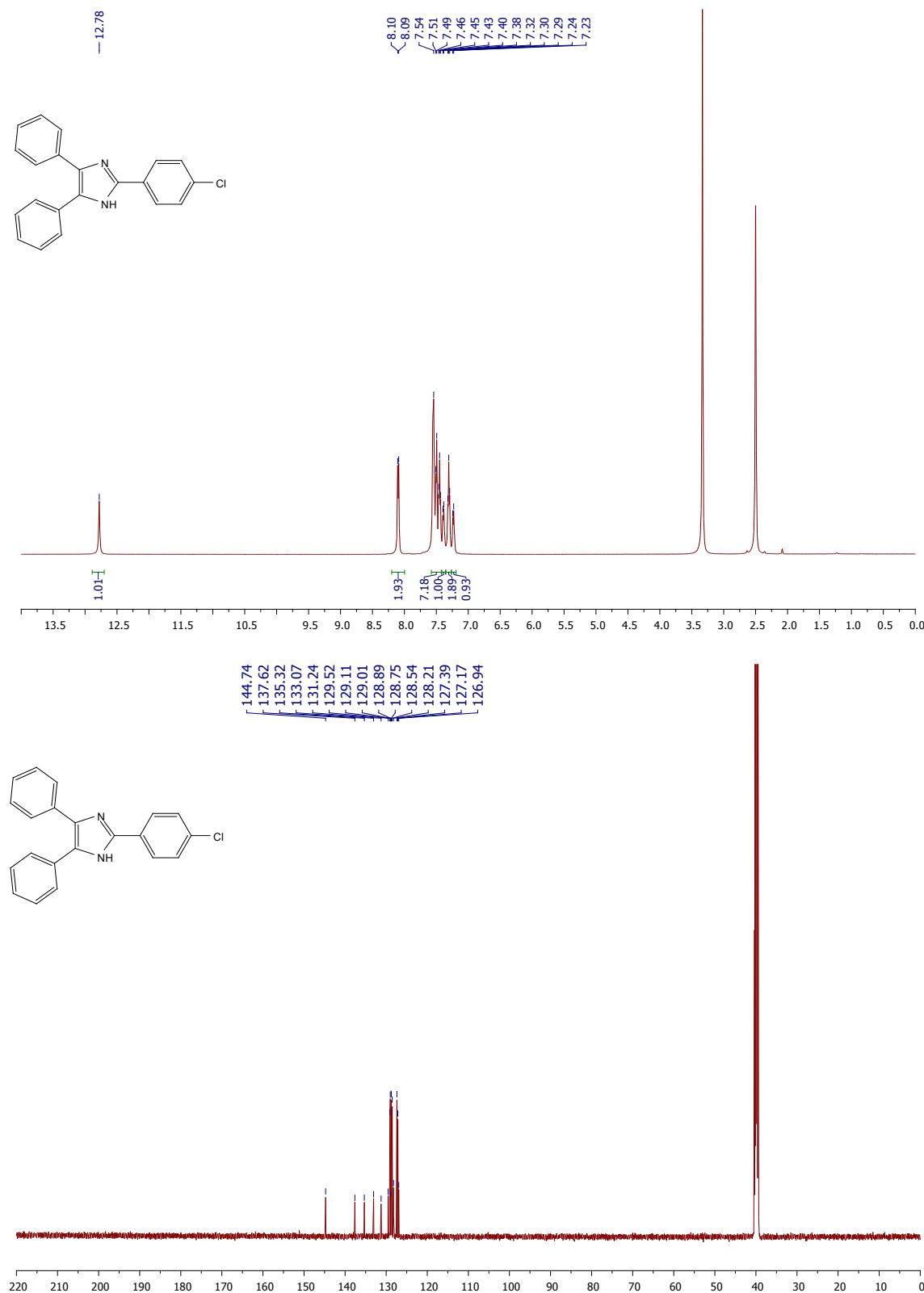


Figure S9. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-chlorophenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{21}\text{H}_{15}\text{ClN}_2$, 1i)

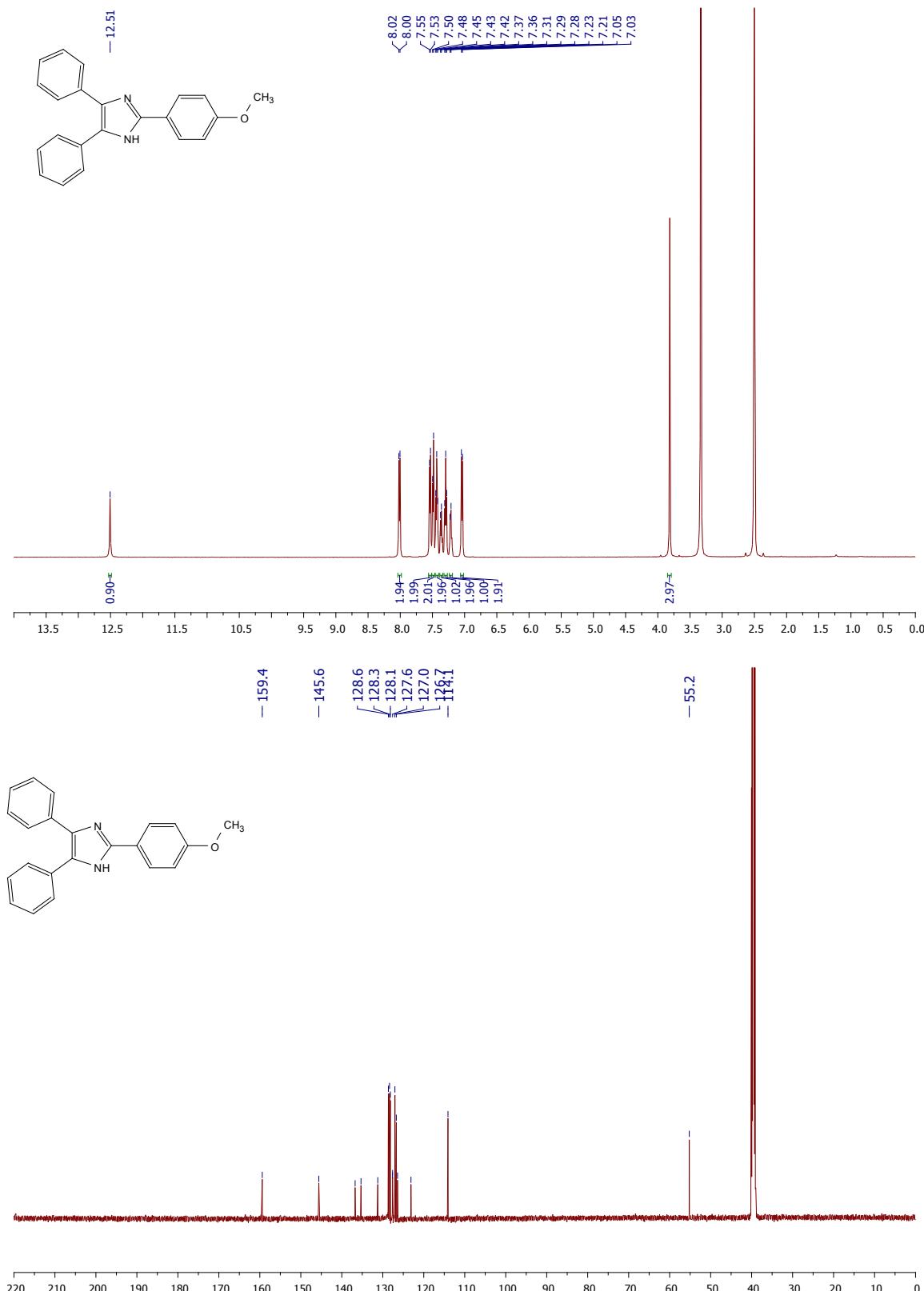


Figure S10. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-methoxyphenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{22}\text{H}_{18}\text{N}_2\text{O}$, 1k)

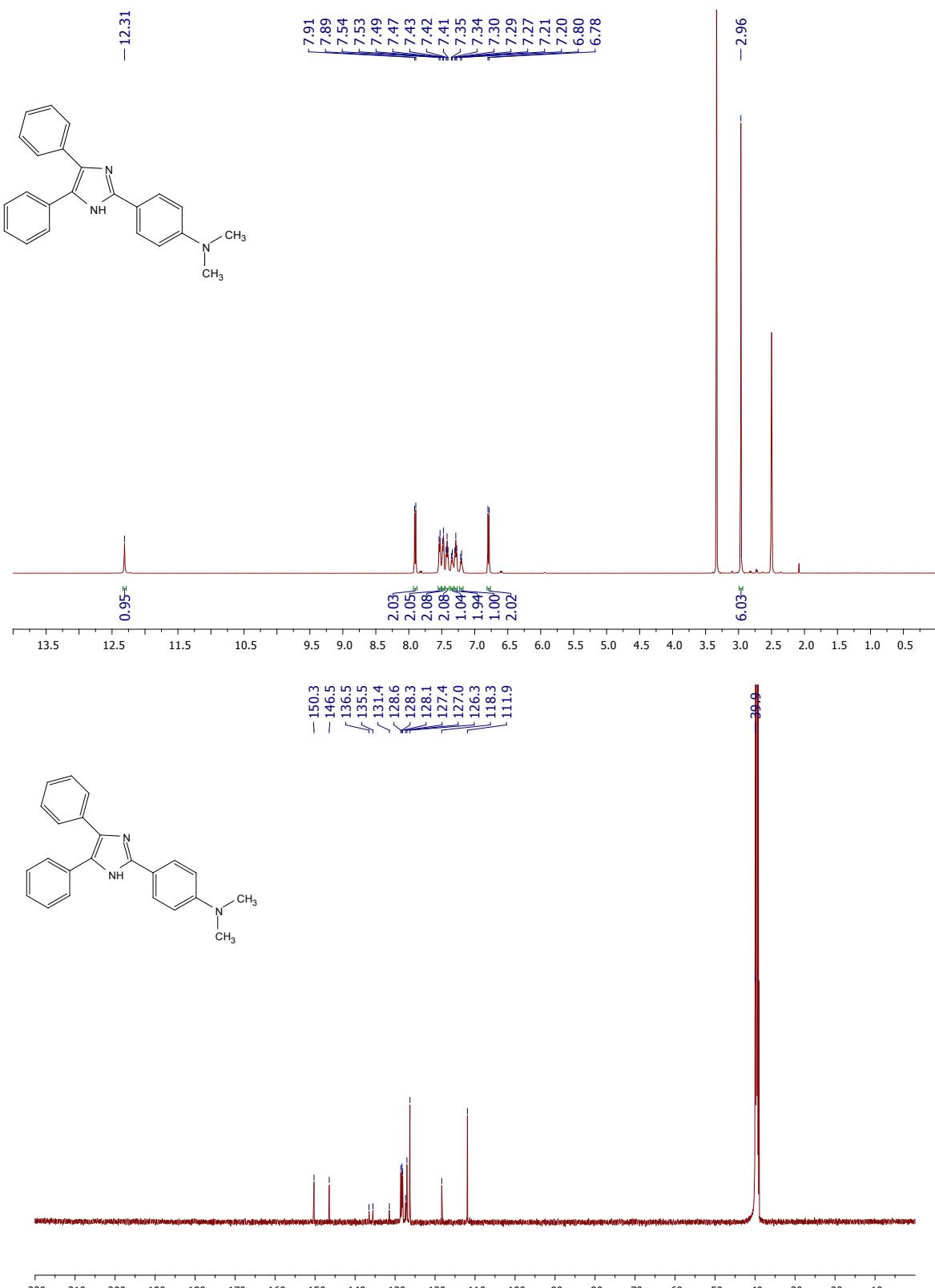


Figure S11. ^1H (top) and ^{13}C (bottom) NMR spectra of 4-(4,5-diphenyl-1*H*-imidazol-2-yl)-*N,N*-dimethylaniline ($\text{C}_{23}\text{H}_{21}\text{N}_3$, **1l)**

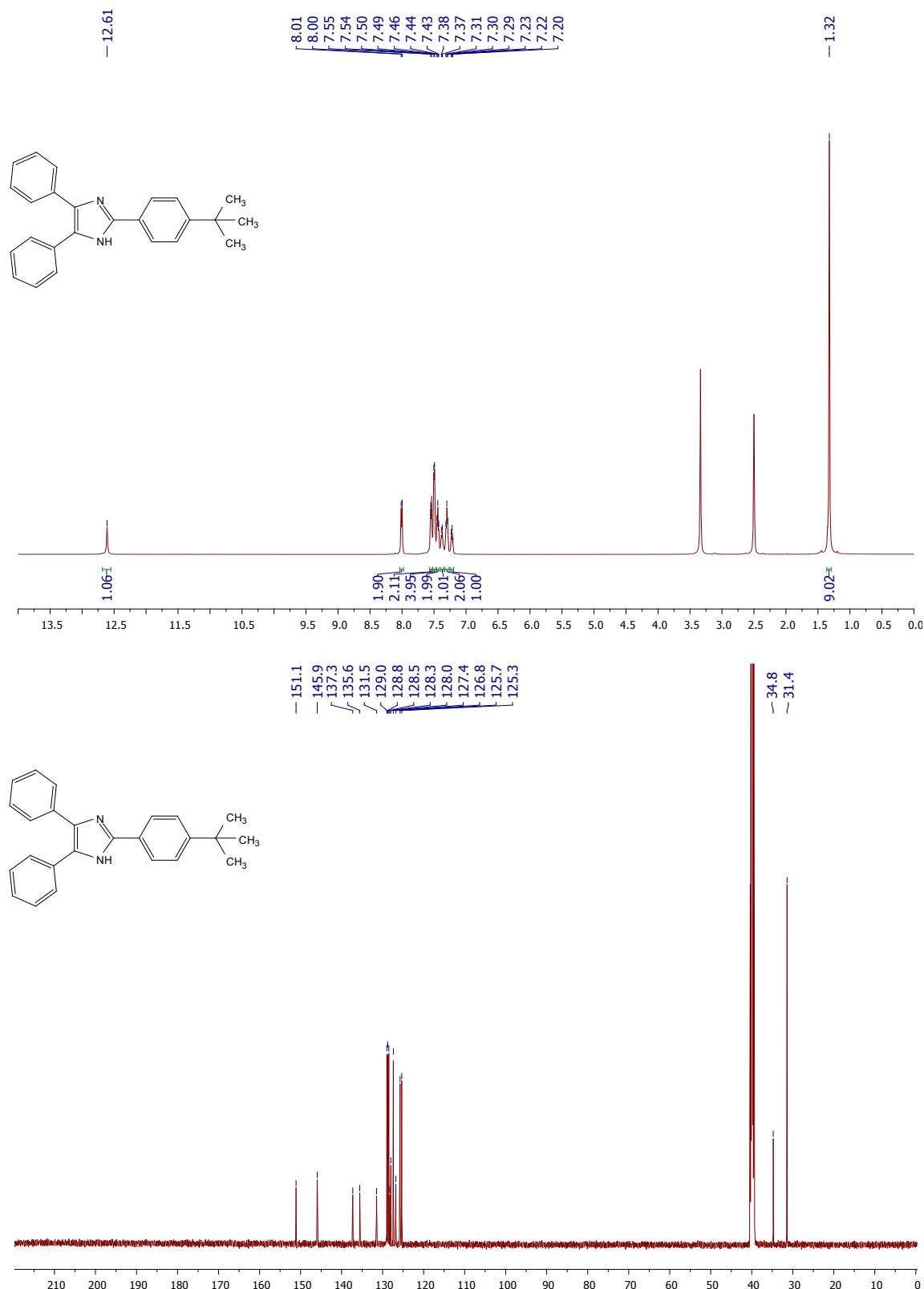


Figure S12. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-(*tert*-butyl)phenyl)-4,5-diphenyl-1*H*-imidazole ($\text{C}_{24}\text{H}_{22}\text{N}_2$, 1m)

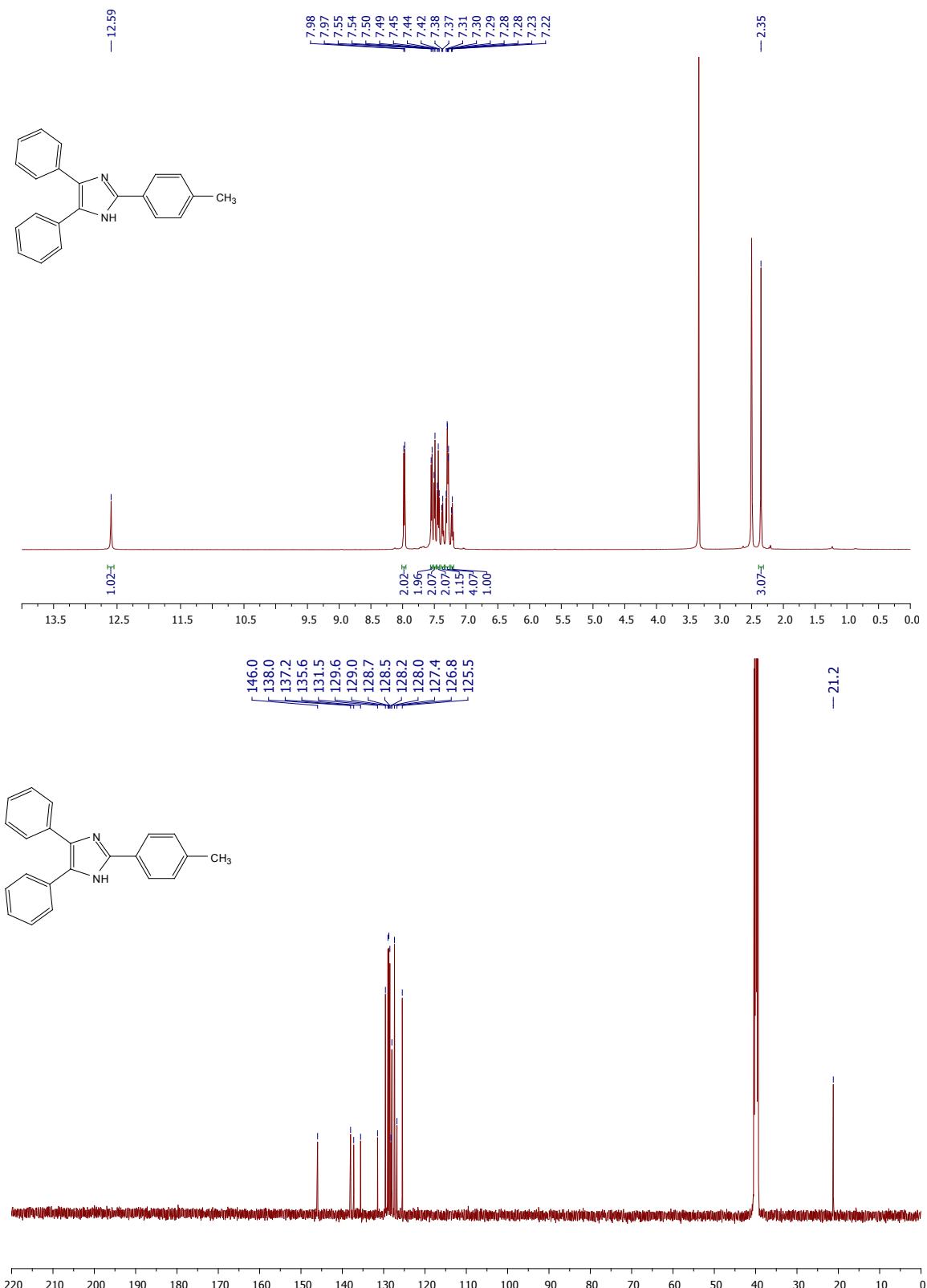


Figure S13. ^1H (top) and ^{13}C (bottom) NMR spectra of 4,5-diphenyl-2-(*p*-tolyl)-1*H*-imidazole ($\text{C}_{22}\text{H}_{18}\text{N}_2$, 1n)

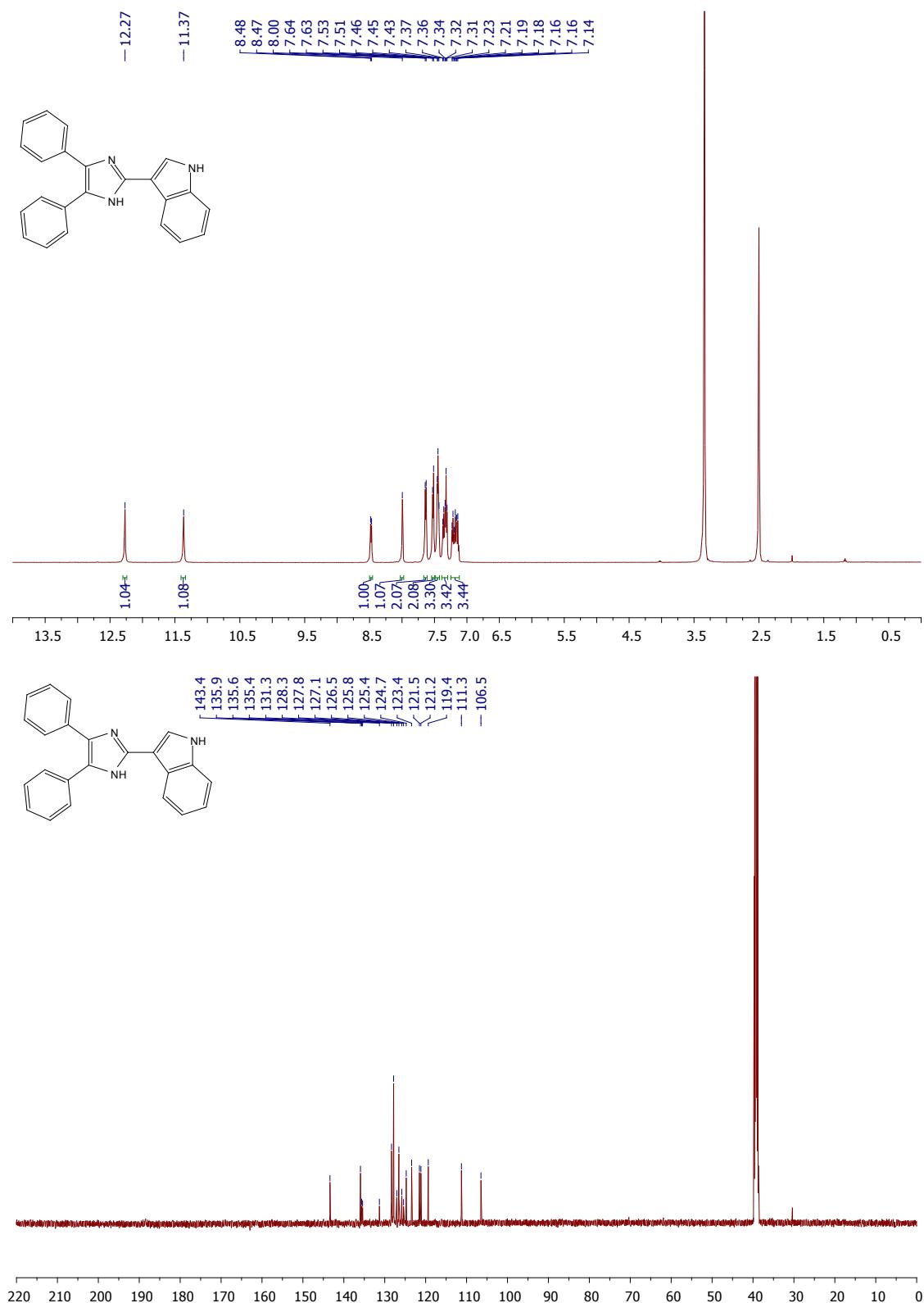
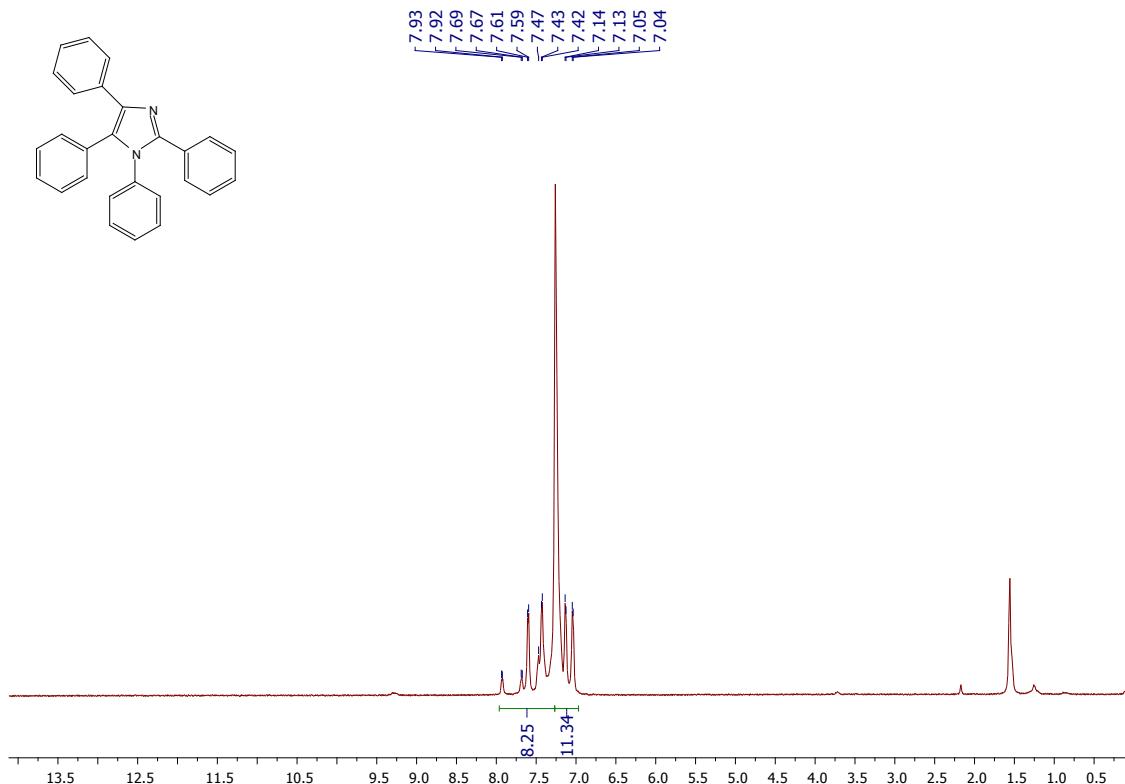


Figure S14. ^1H (top) and ^{13}C (bottom) NMR spectra of 3-(4,5-diphenyl-1*H*-imidazol-2-yl)-1*H*-indole ($\text{C}_{23}\text{H}_{17}\text{N}_3$, **1o)**



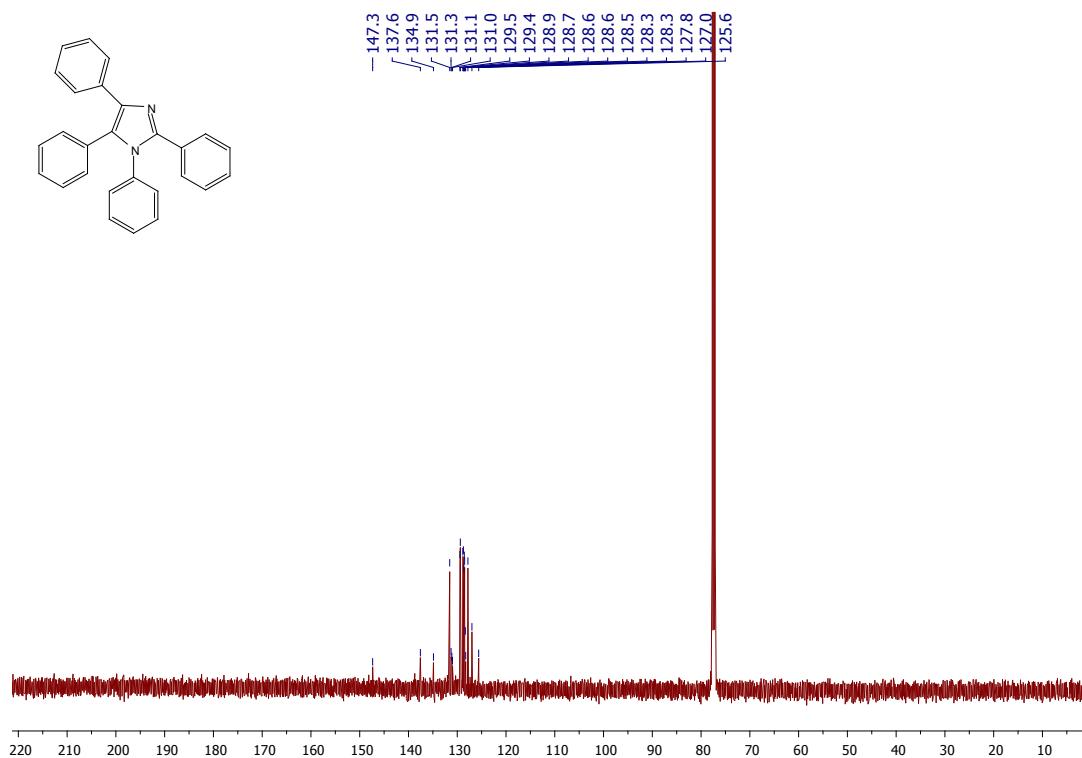
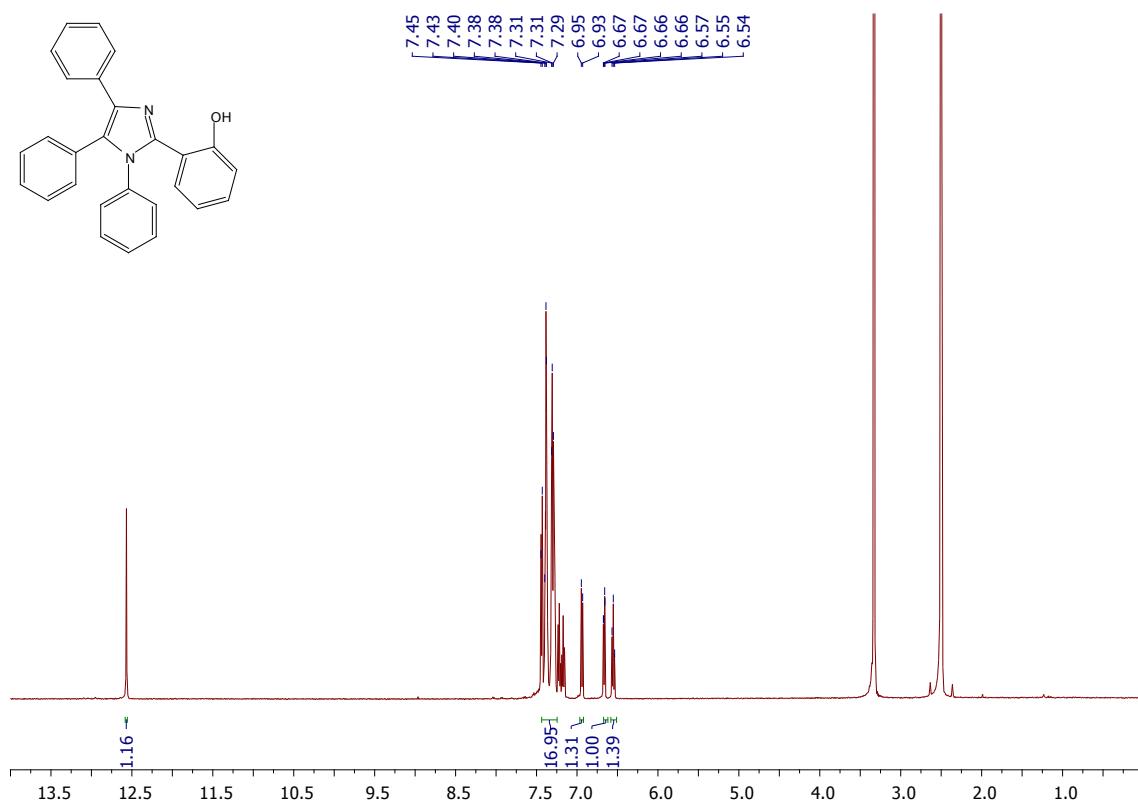


Figure S15. 1H (top) and ^{13}C (bottom) NMR spectra of 1,2,4,5-tetraphenyl imidazole ($C_{27}H_{20}N_2$, **2a**)



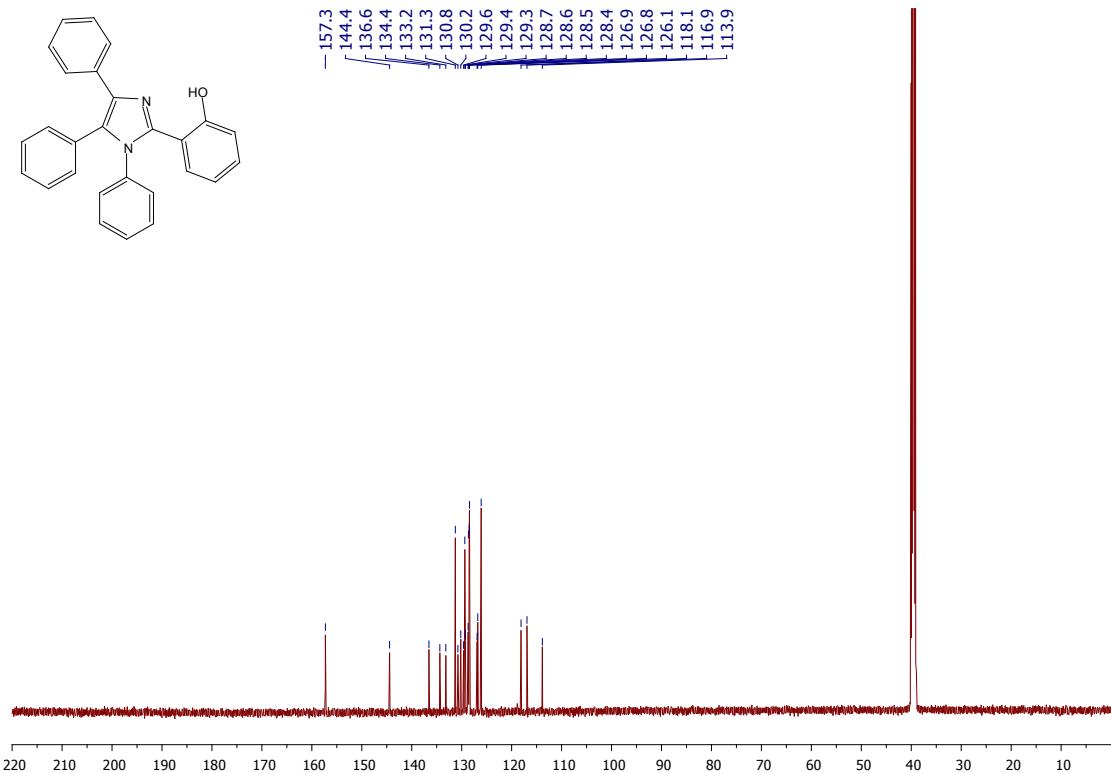
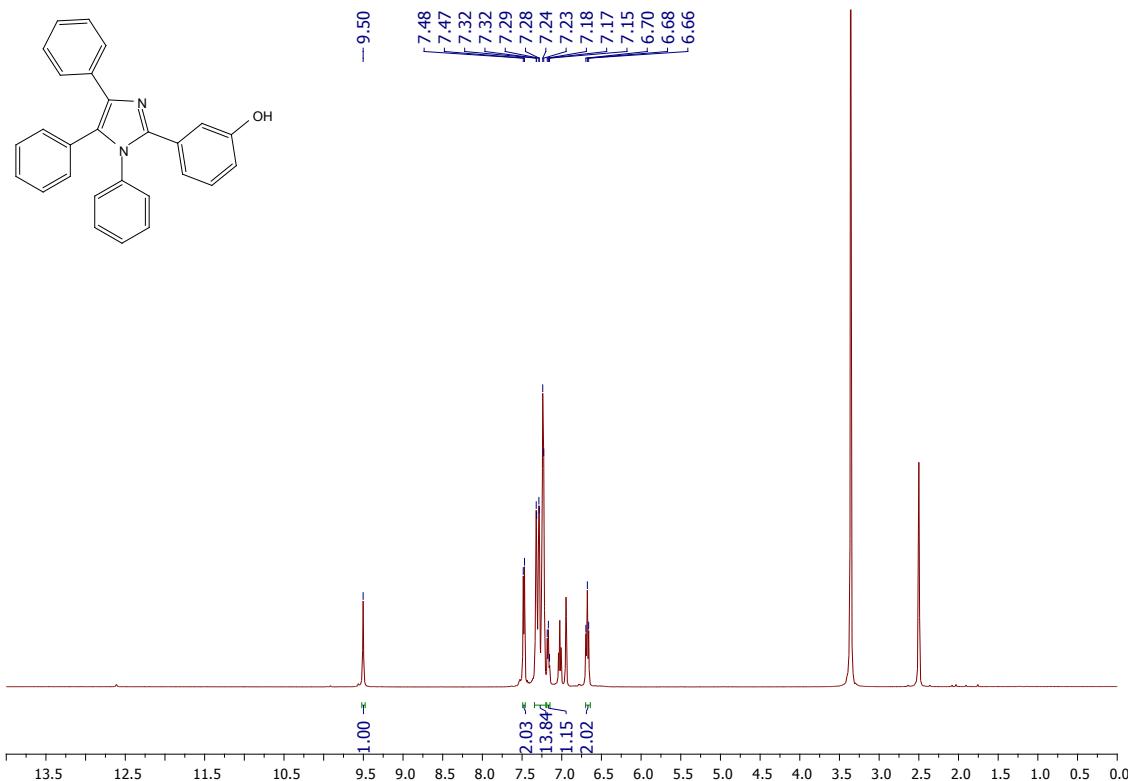


Figure S16. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(2-hydroxyphenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{20}N_2O$, 2b)



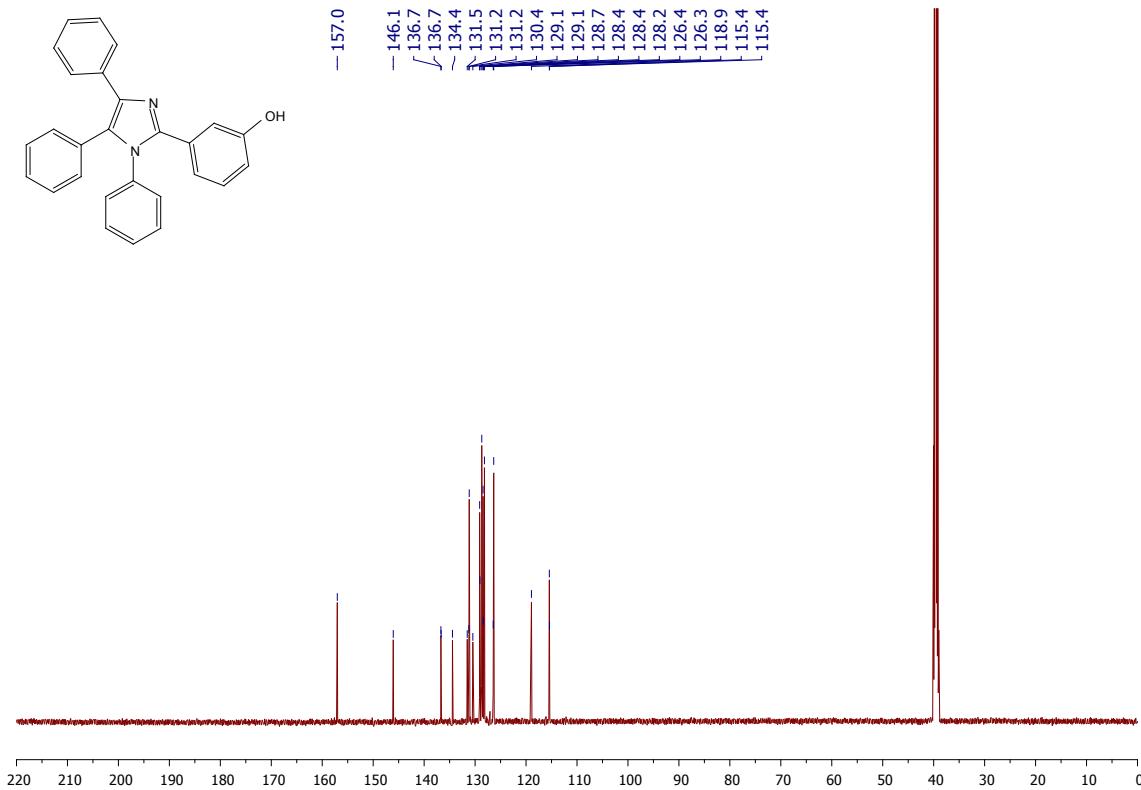
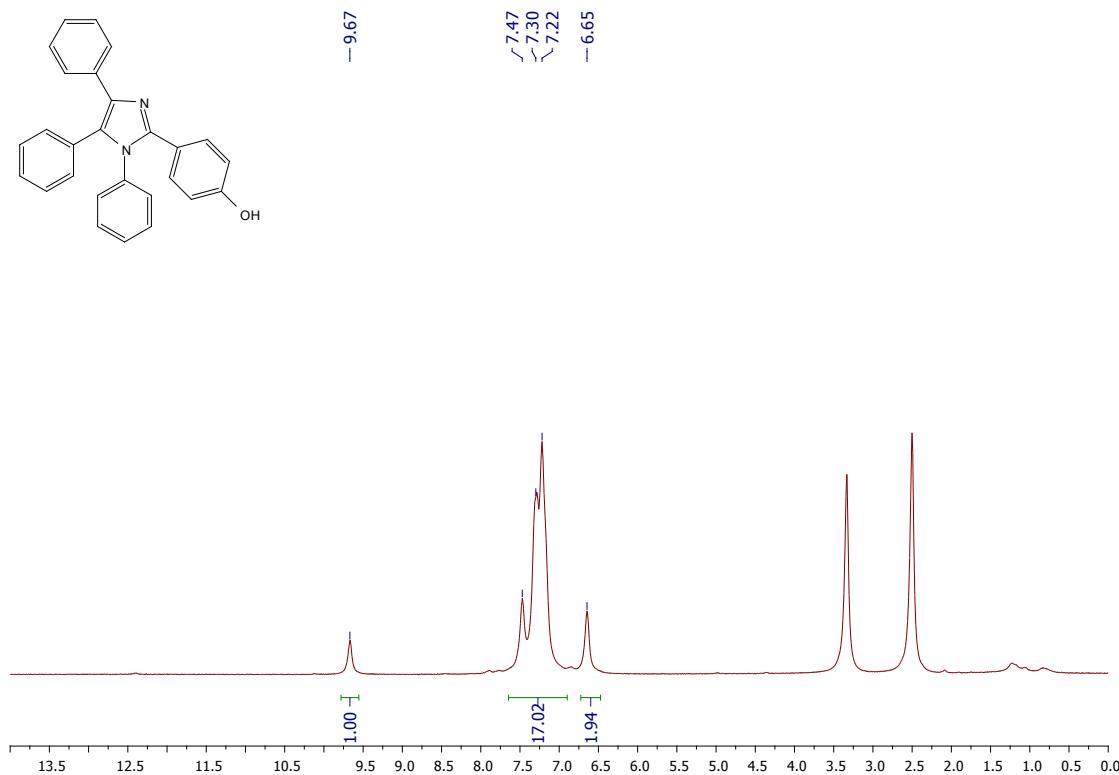


Figure S17. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(3-hydroxyphenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{20}N_2O$, **2c**)



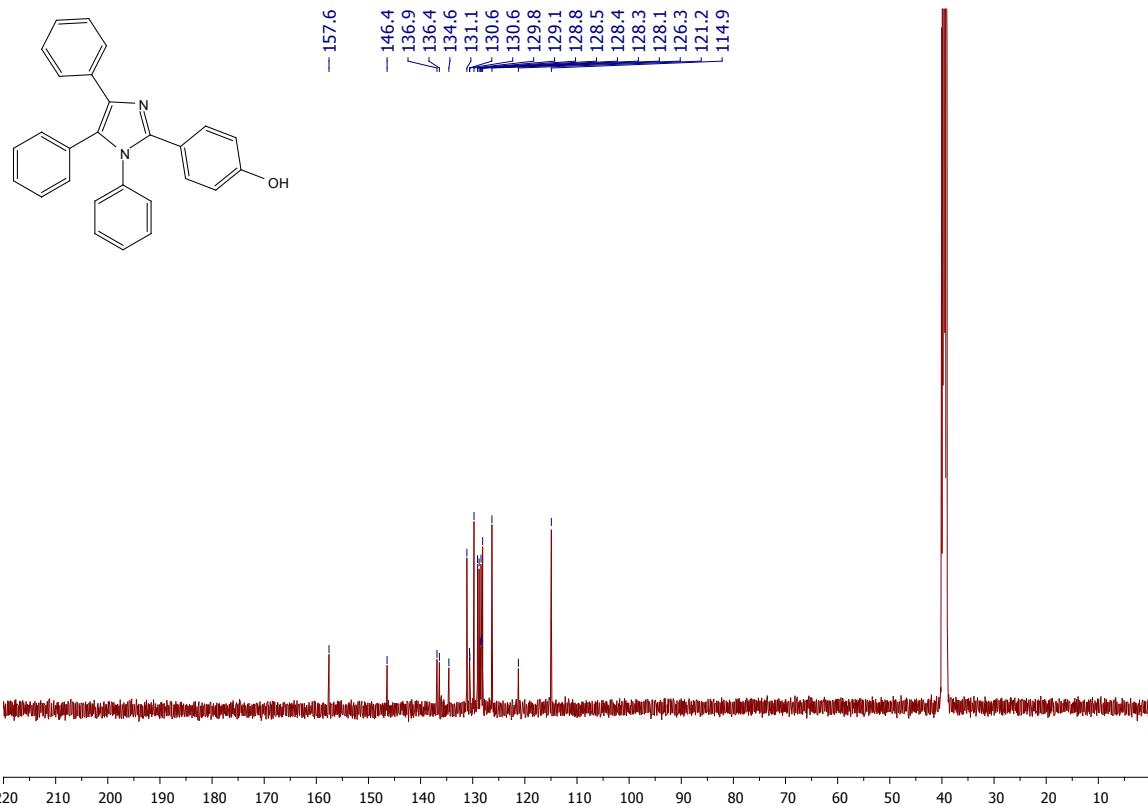
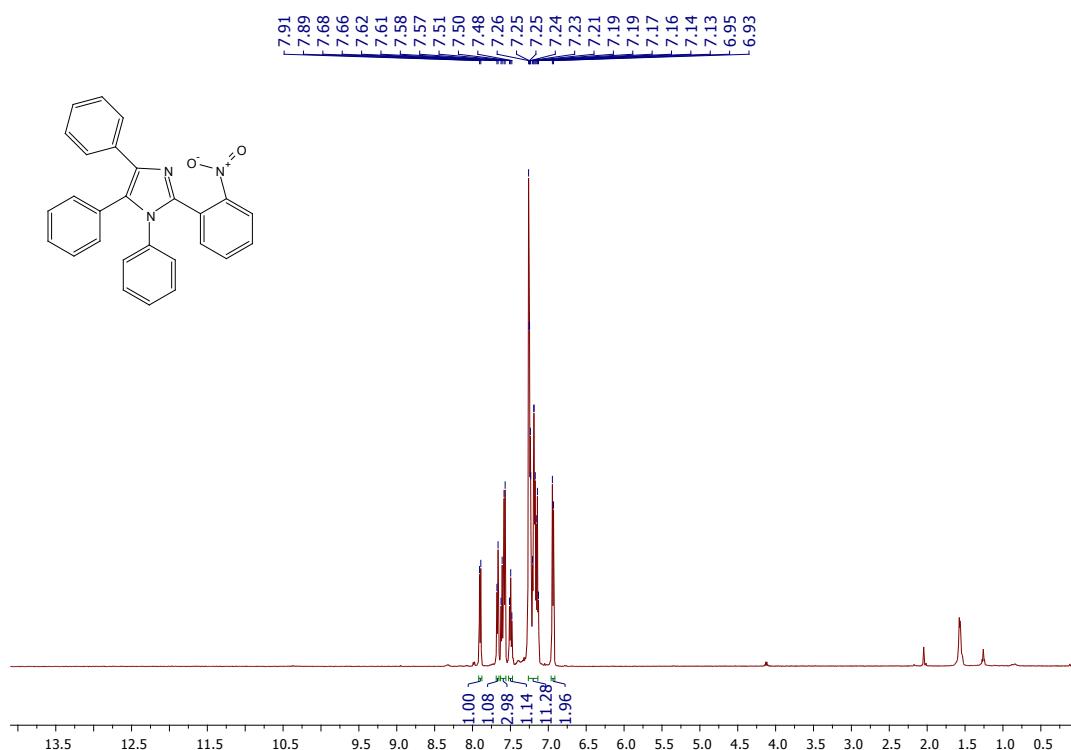


Figure S18. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(4-hydroxyphenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{20}N_2O$, 2d)



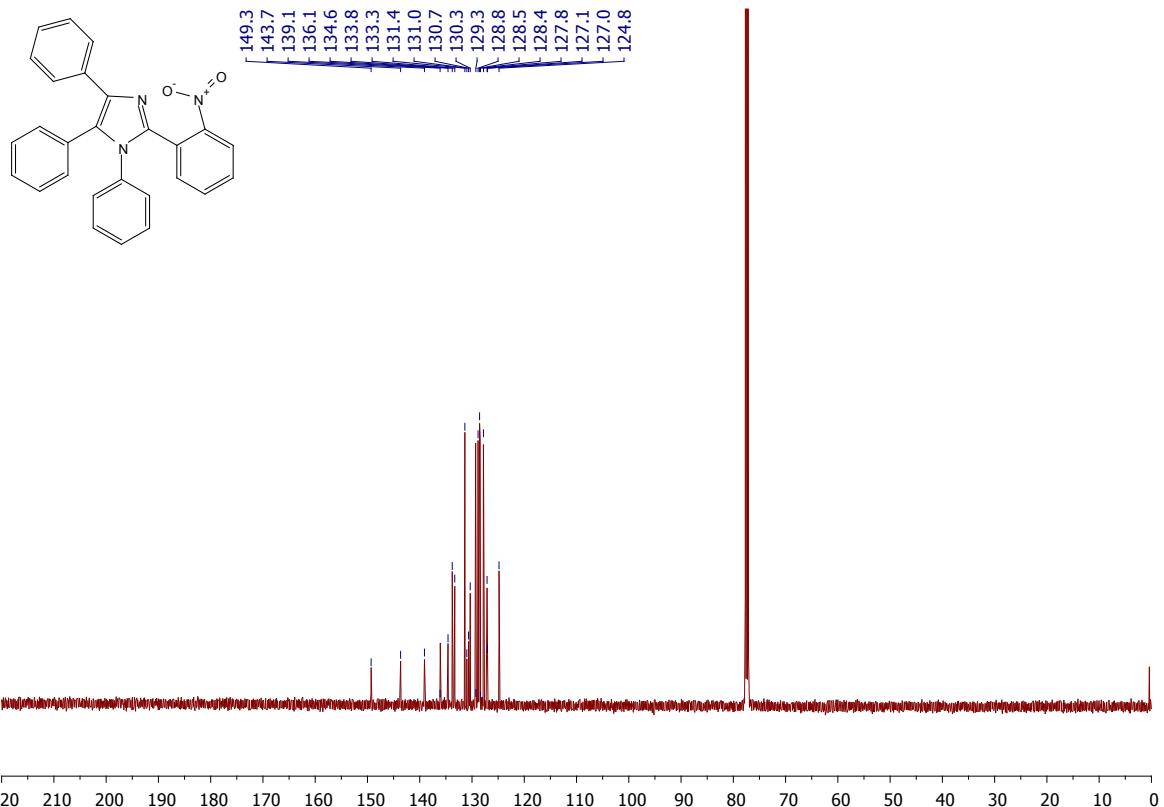
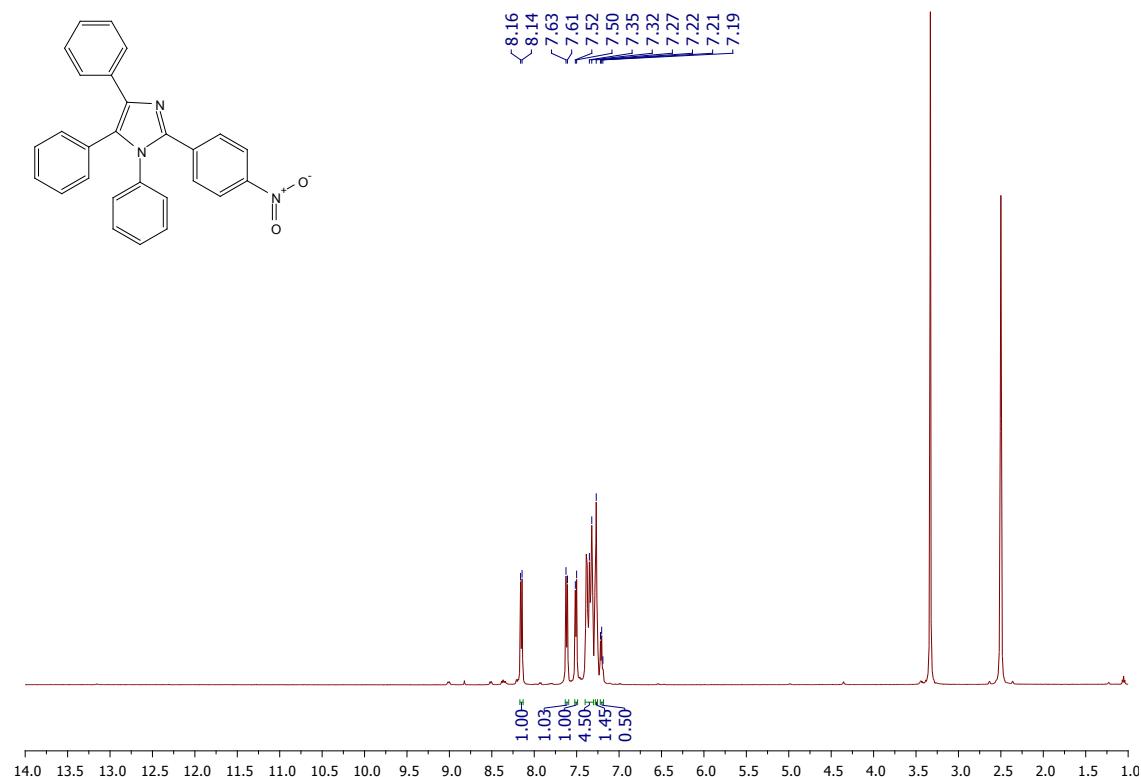


Figure S19. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(2-nitrophenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{19}N_3O_2$, 2e)



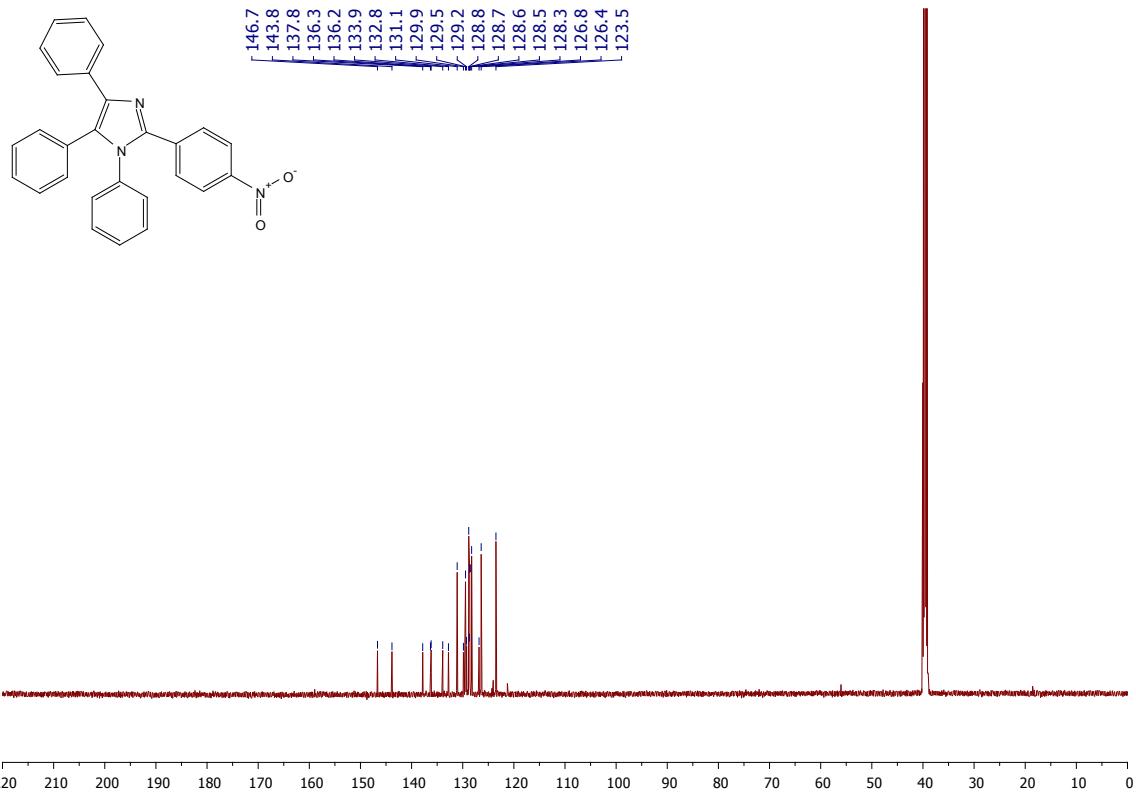
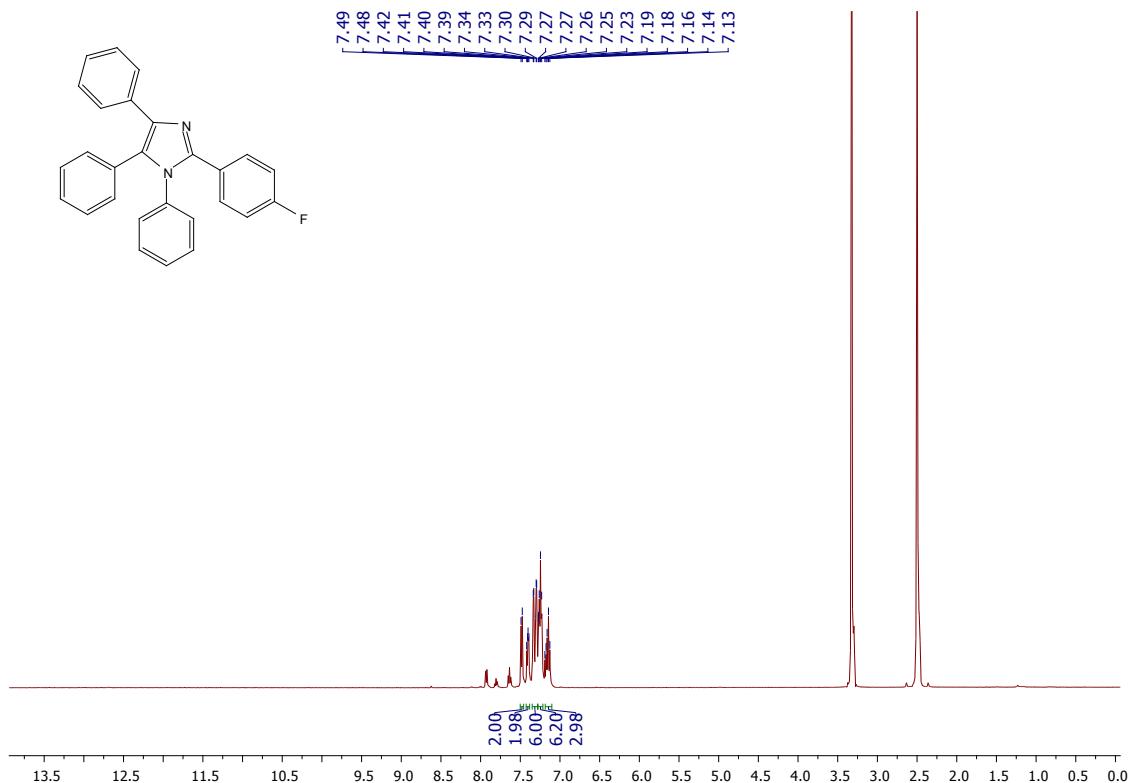
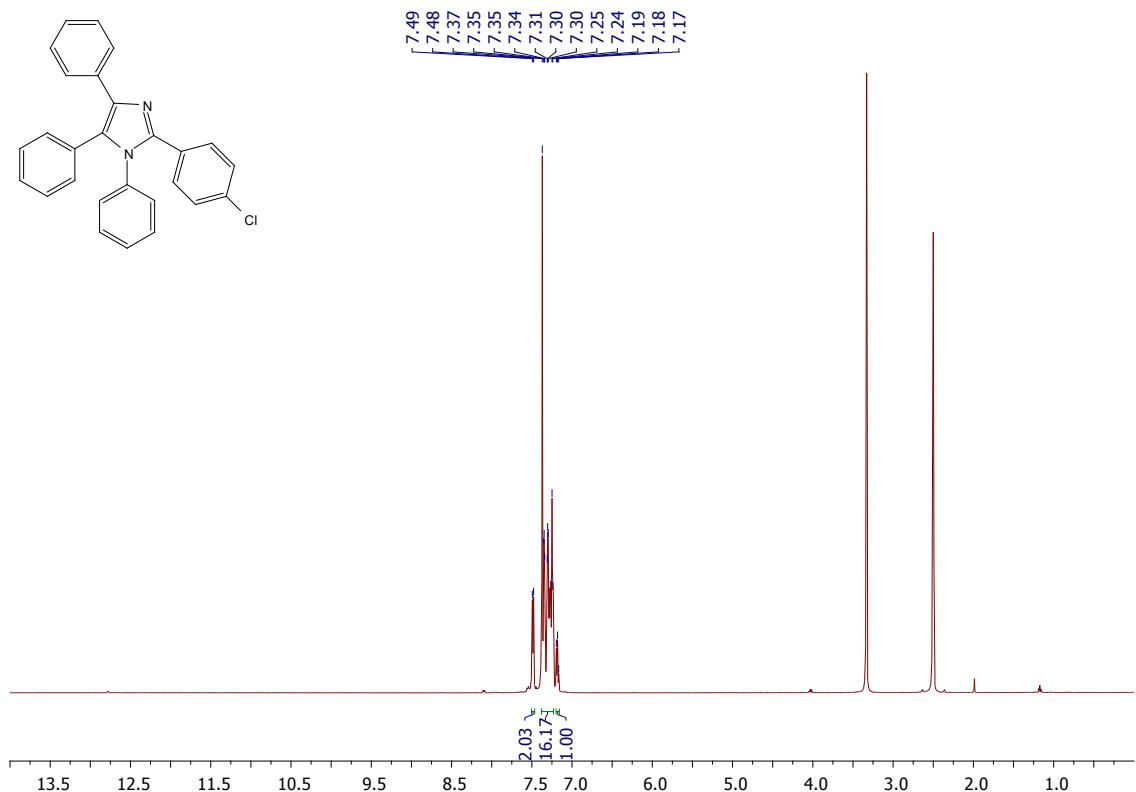
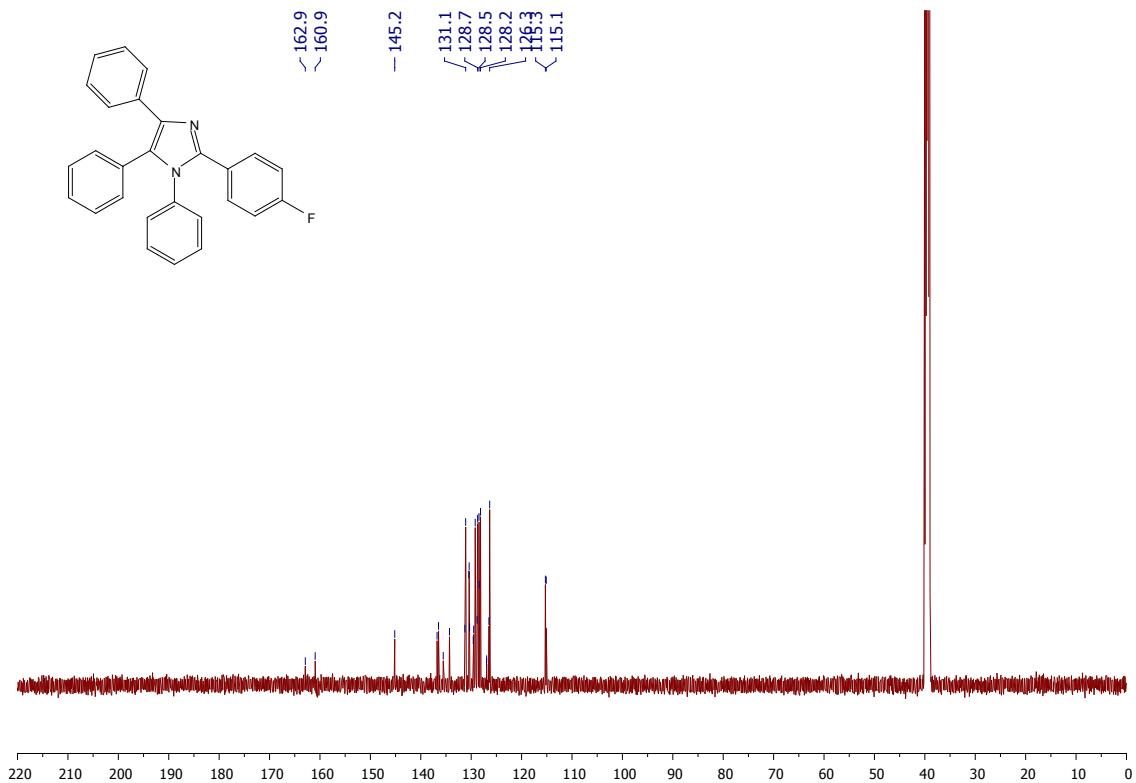


Figure S20. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(4-nitrophenyl)-1,4,5-triphenyl imidazole ($C_{27}H_{19}N_3O_2$, 2f)





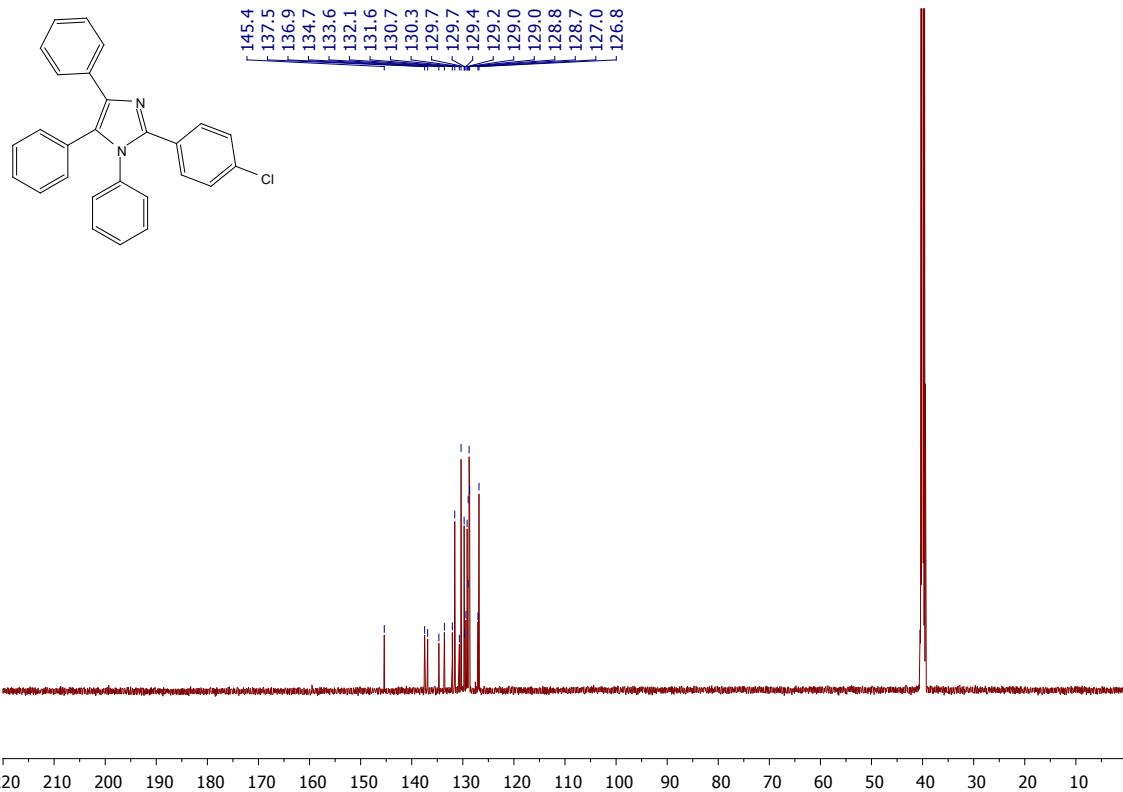
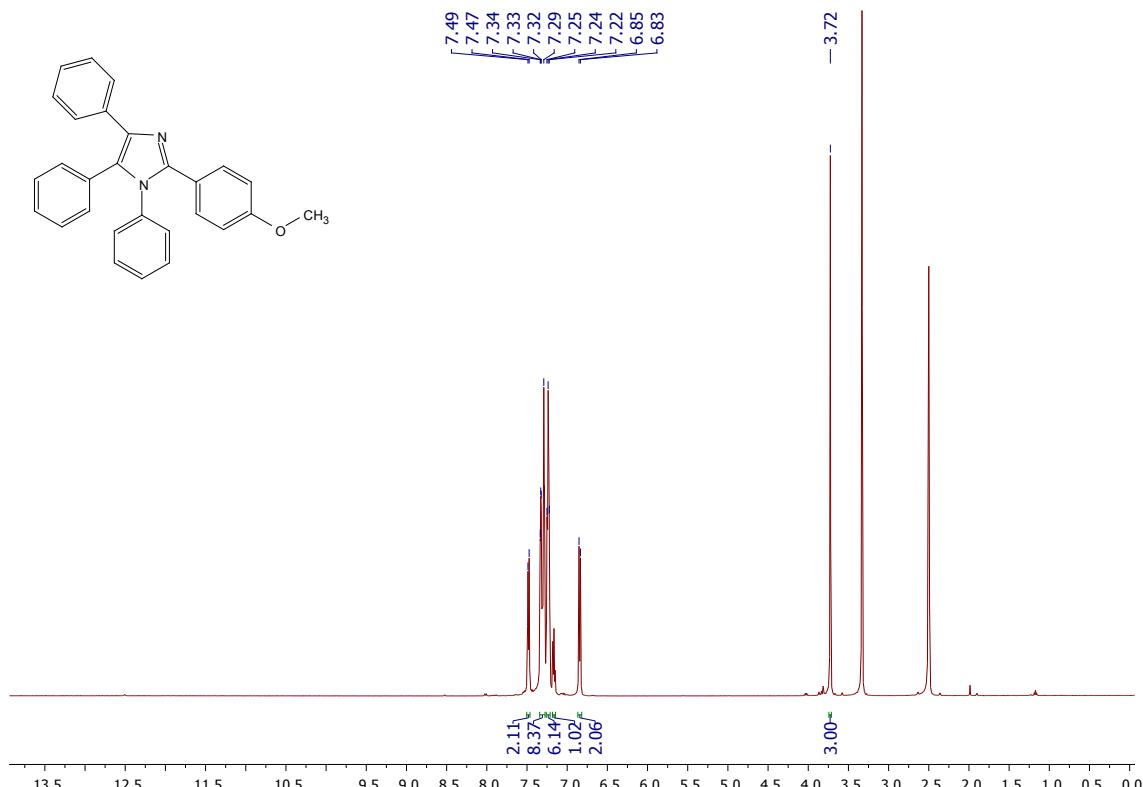


Figure S22. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-chlorophenyl)-1,4,5-triphenyl imidazole ($\text{C}_{27}\text{H}_{19}\text{N}_2\text{Cl}$, 2h)



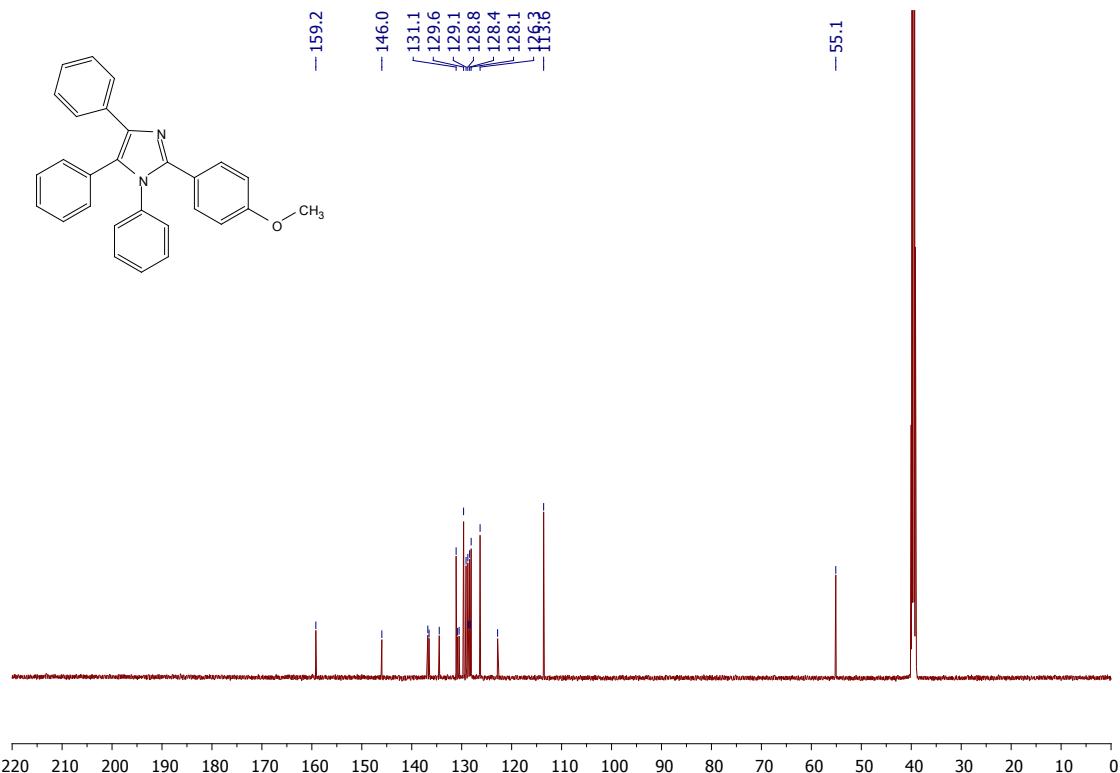
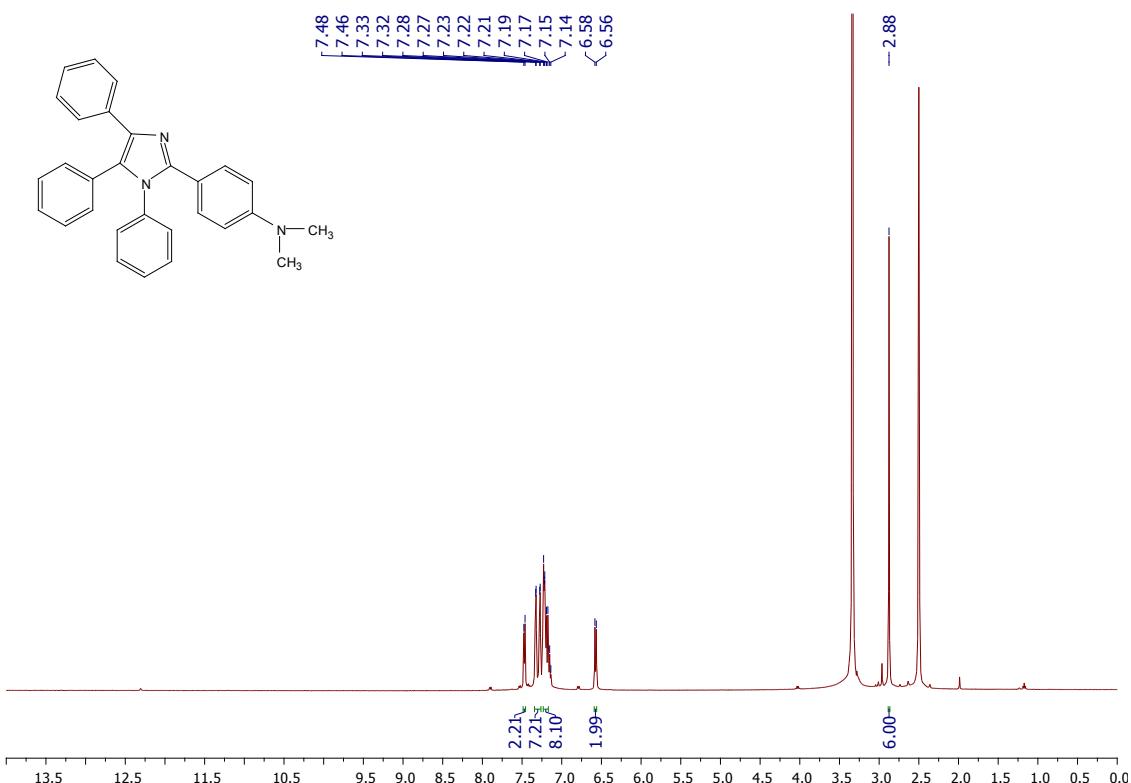


Figure S23. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(4-methoxyphenyl)-1,4,5-triphenyl imidazole ($C_{28}H_{22}N_2O$, **2k**)



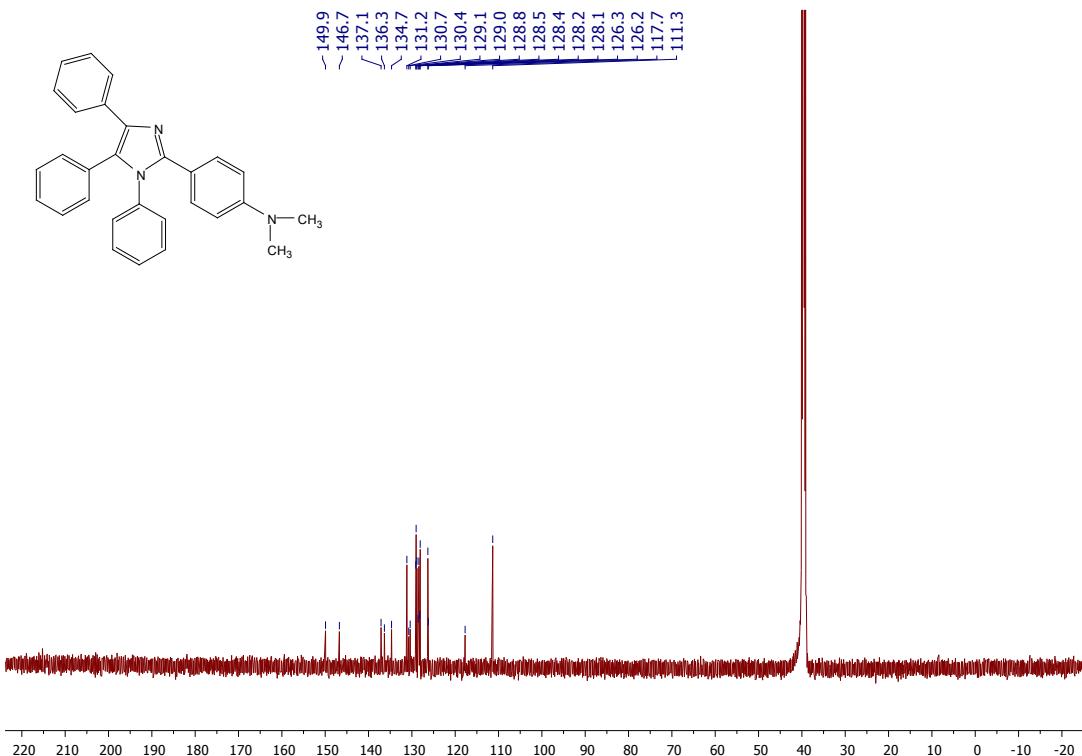
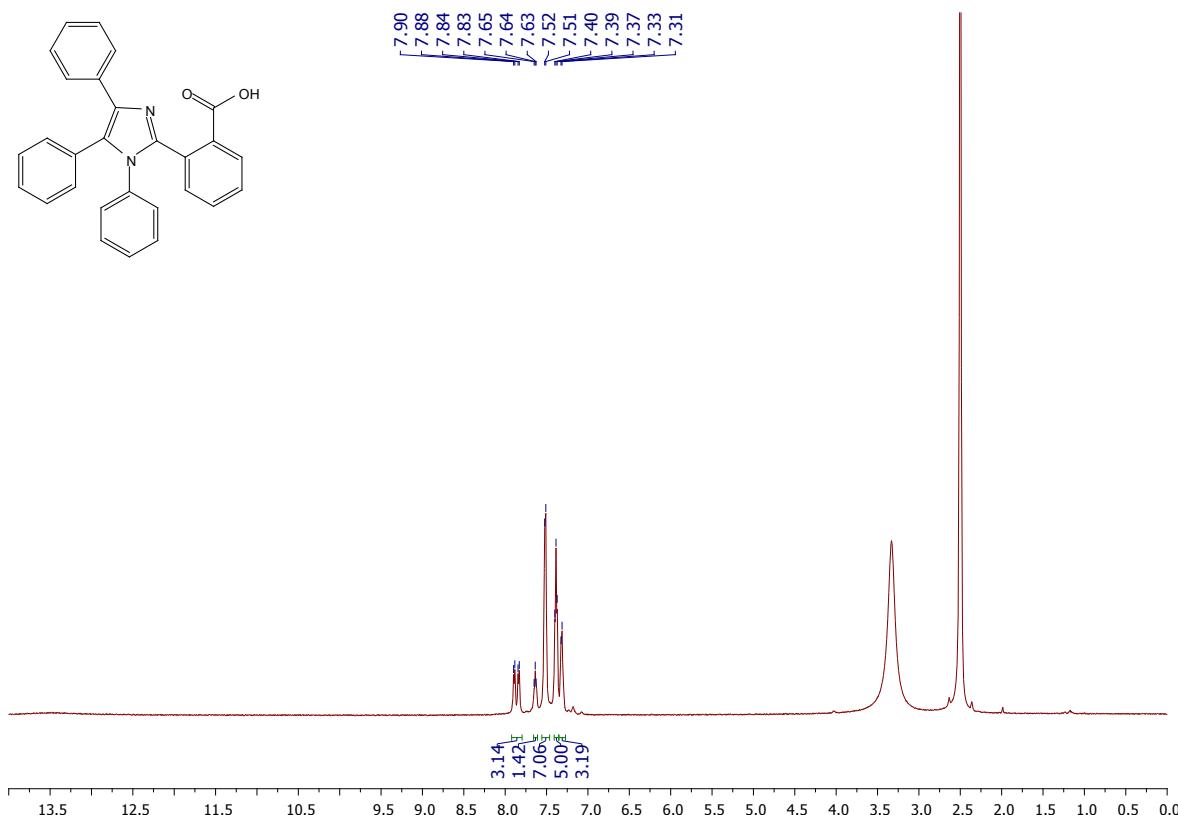


Figure S24. ^1H (top) and ^{13}C (bottom) NMR spectra of 2-(4-N,N-dimethylaminophenyl)-1,4,5-triphenyl imidazole ($\text{C}_{29}\text{H}_{25}\text{N}_3$, 2l)



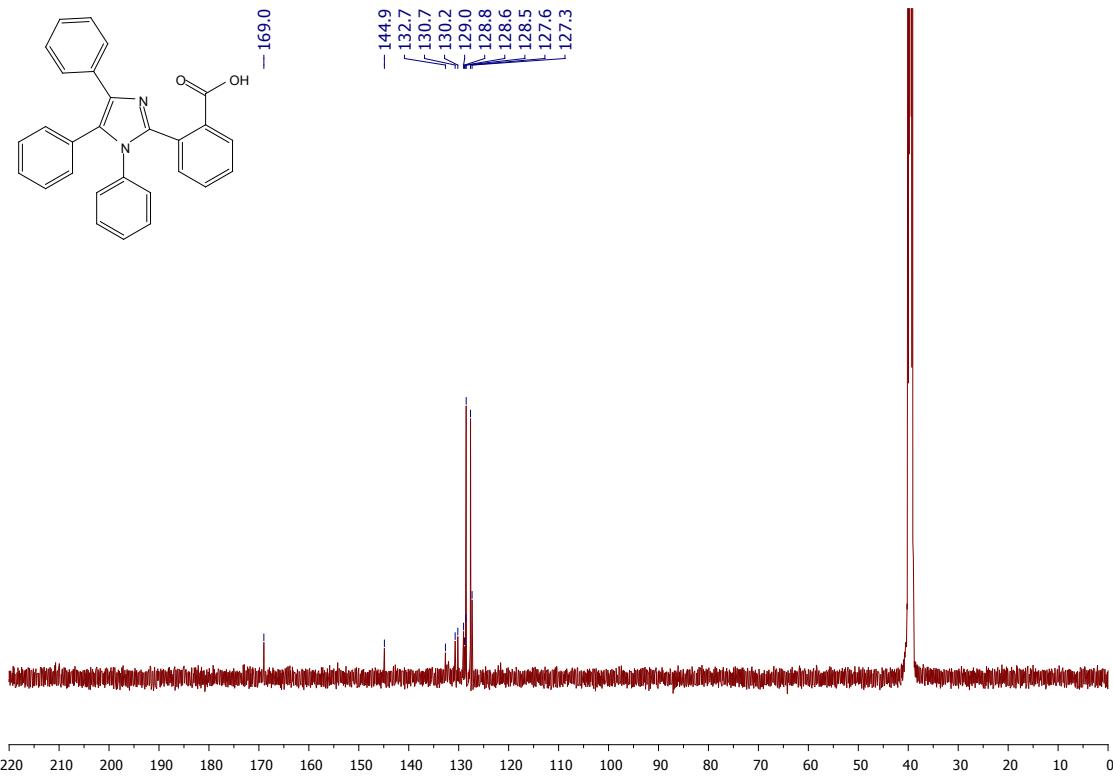
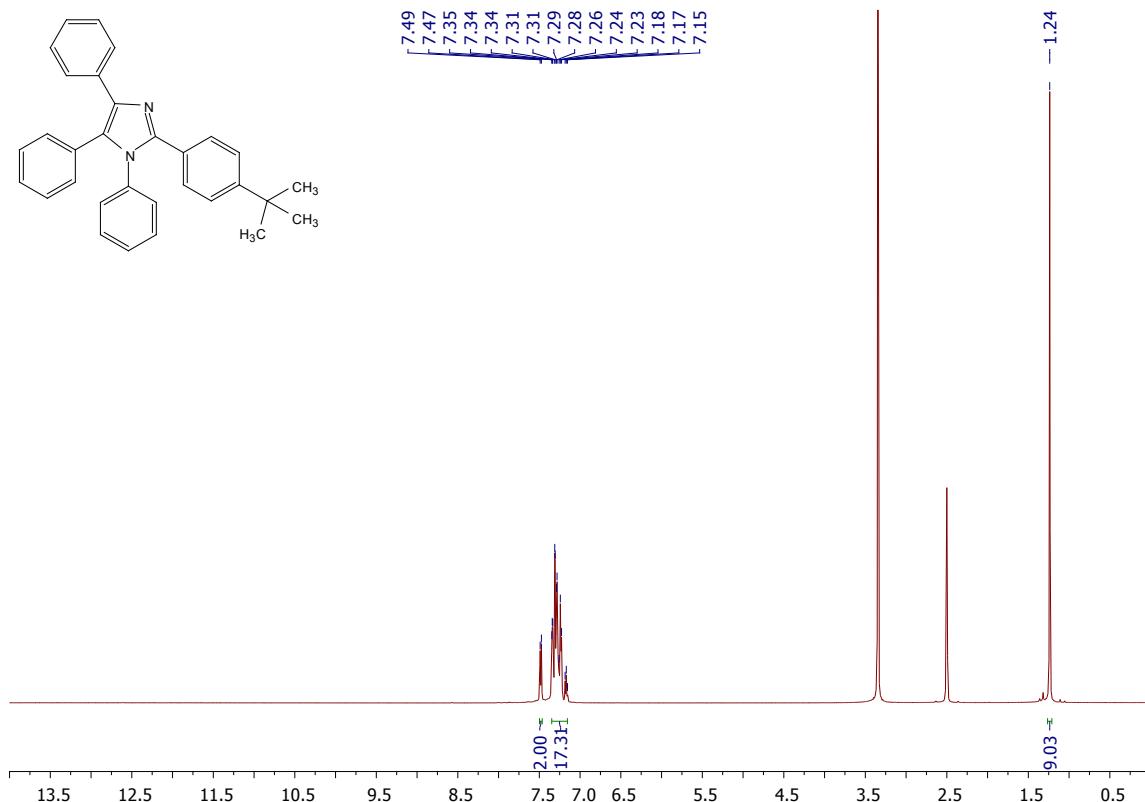


Figure S25. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(2-carboxyphenyl)-1,4,5-triphenyl imidazole ($C_{28}H_{20}N_2O_2$, 2m)



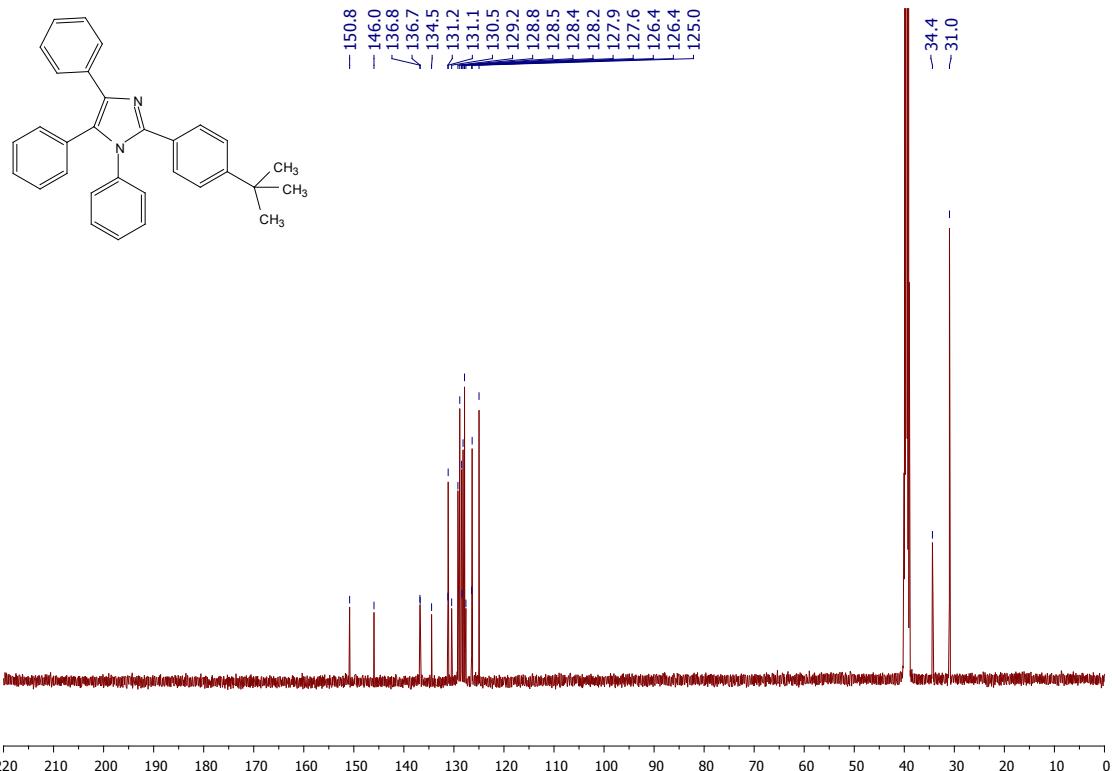
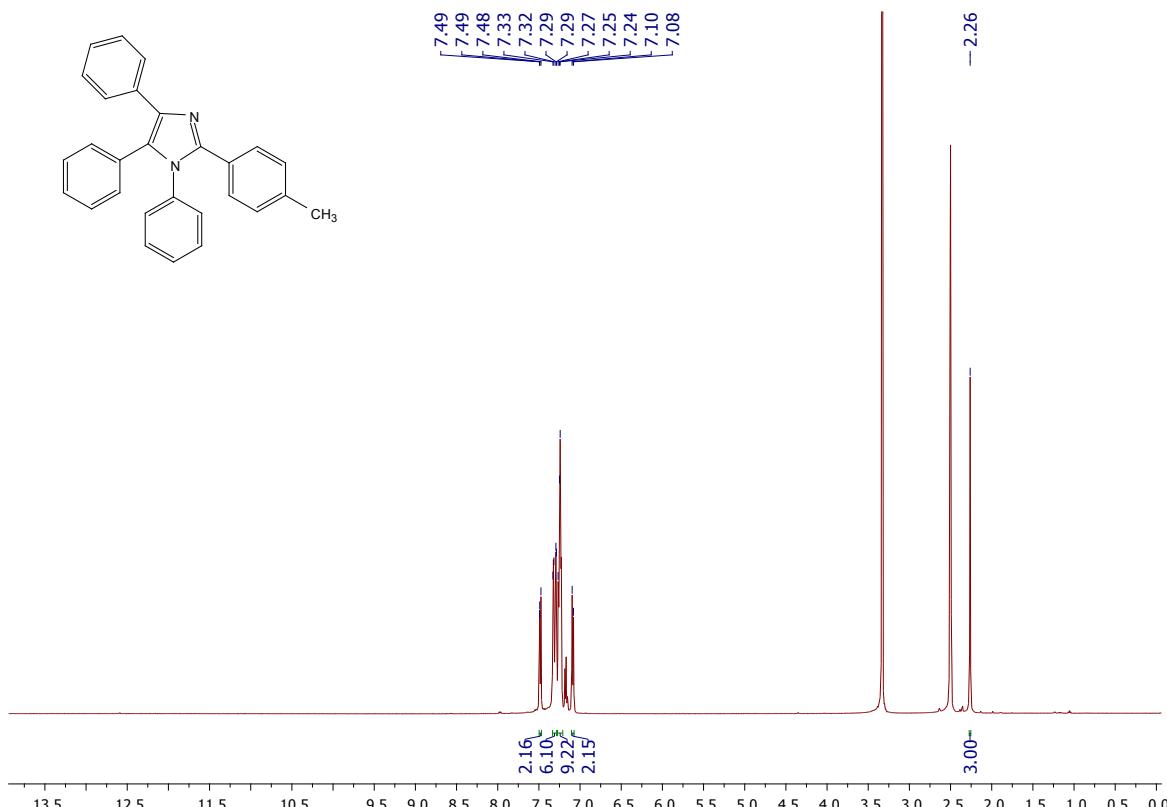


Figure S26. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(4-*tert*-butylphenyl)-1,4,5-triphenyl imidazole ($C_{31}H_{29}N_2$, 2o)



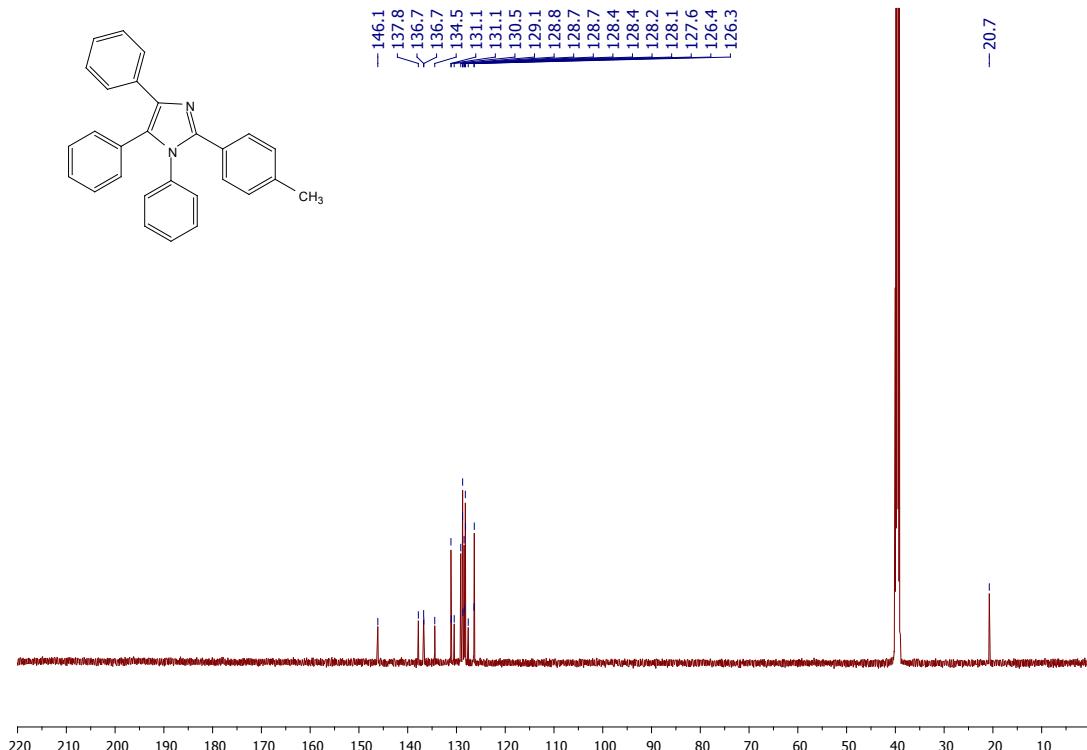
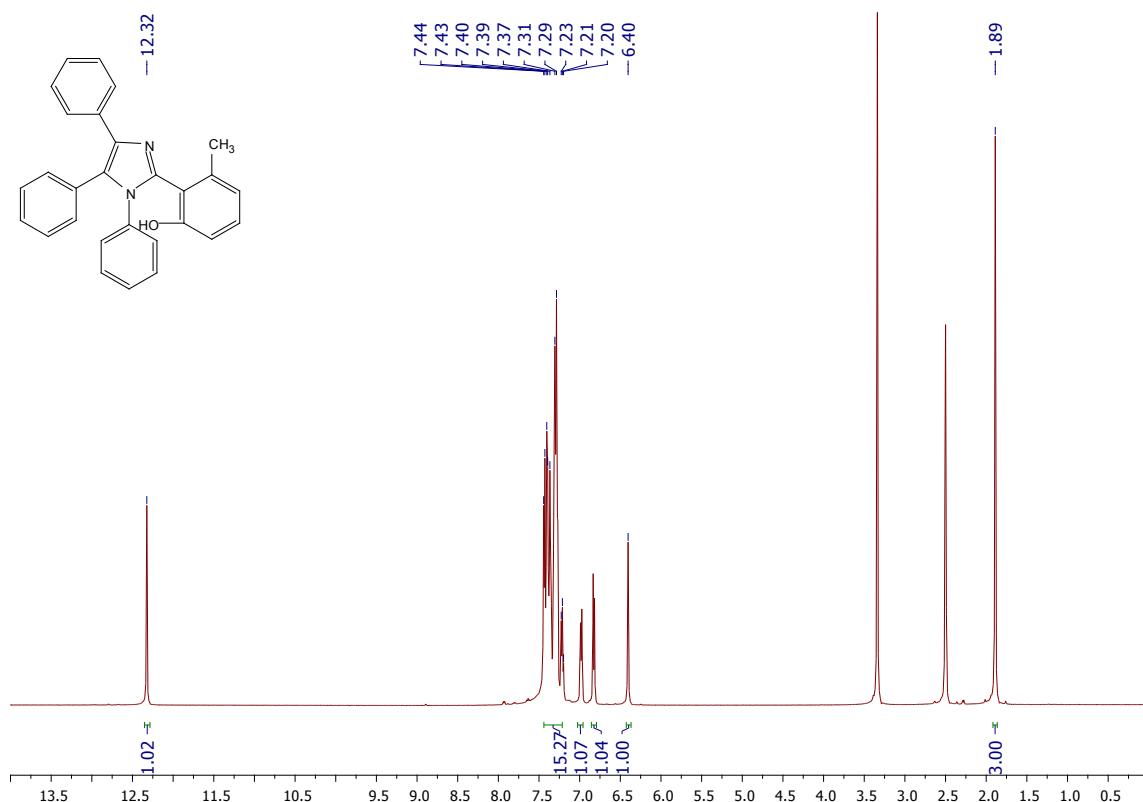


Figure S27. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(4-methylphenyl)-1,4,5-triphenylimidazole ($C_{28}H_{22}N_2$, 2p)



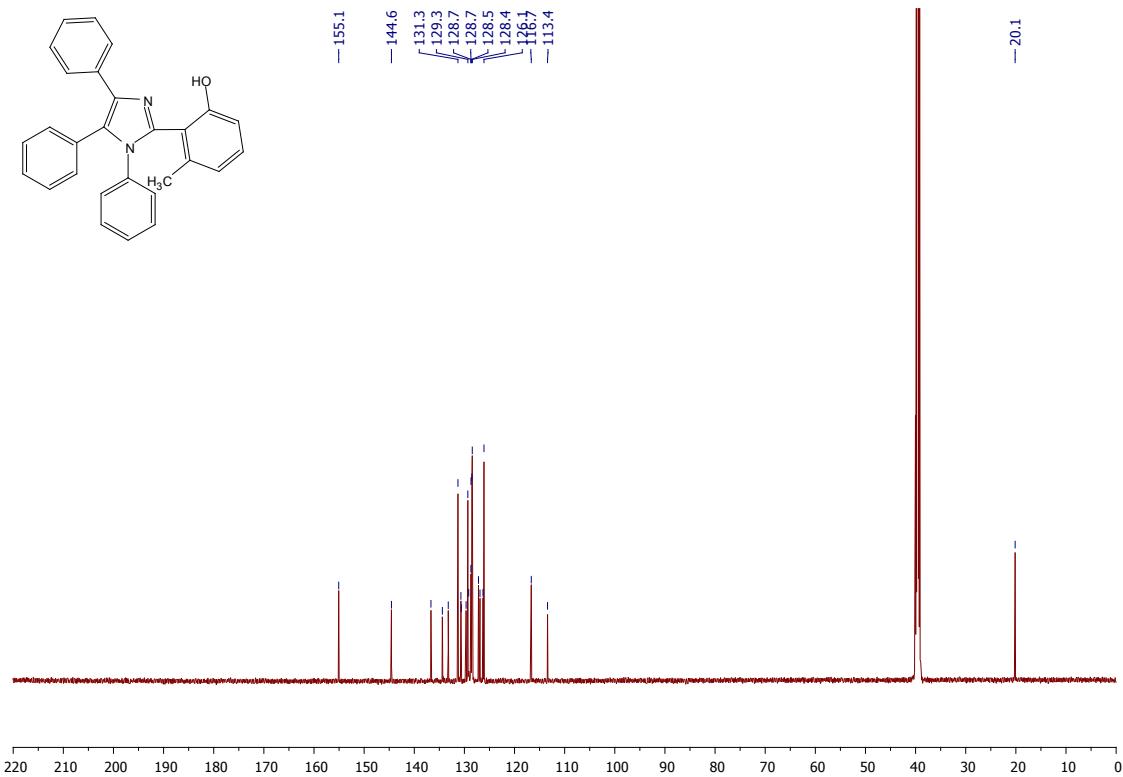


Figure S28. ¹H (top) and ¹³C (bottom) NMR spectra of 2-(2-hydroxy-5-methylphenyl)-1,4,5-triphenyl imidazole ($C_{28}H_{23}N_2O$, 2q)

Section S5. References

1. Smith, E. L. Abbott, A. P. Ryder, K. S., Deep eutectic solvents (DESs) and their applications. *Chem. Rev.* **2014**, *114*, 11060-11082.
2. Clarke, C. J. Tu, W. C. Levers, O. Brohl, A. Hallett, J. P., Green and Sustainable Solvents in Chemical Processes. *Chem. Rev.* **2018**, *118*, 747-800.
3. Liu, B. Fu, W. Lu, X. Zhou, Q. Zhang, S., Lewis Acid–Base Synergistic Catalysis for Polyethylene Terephthalate Degradation by 1,3-Dimethylurea/Zn(OAc)₂ Deep Eutectic Solvent. *ACS Sus. Chem. Eng.* **2018**, *7*, 3292-3300.
4. Tomé, L. I. N. Baião, V. da Silva, W. Brett, C. M. A., Deep eutectic solvents for the production and application of new materials. *Appl. Mater. Today* **2018**, *10*, 30-50.
5. Hong, S. Lian, H. Sun, X. Pan, D. Carranza, A. Pojman, J. A. Mota-Morales, J. D., Zinc-based deep eutectic solvent-mediated hydroxylation and demethoxylation of lignin for the production of wood adhesive. *RSC Adv.* **2016**, *6*, 89599-89608.
6. Nguyen, H. T. Tran, P. H., An extremely efficient and green method for the acylation of secondary alcohols, phenols and naphthols with a deep eutectic solvent as the catalyst. *RSC Adv.* **2016**, *6*, 98365-98368.
7. Tran, P. H. Nguyen, H. T. Hansen, P. E. Le, T. N., An efficient and green method for regio- and chemo-selective Friedel-Crafts acylations using a deep eutectic solvent ([CholineCl][ZnCl₂]₃). *RSC Adv.* **2016**, *6*, 37031-37038.
8. Nguyen, H. T. Chau, D. K. N. Tran, P. H., A green and efficient method for the synthesis of pyrroles using a deep eutectic solvent ([CholineCl][ZnCl₂]₃) under solvent-free sonication. *New J. Chem.* **2017**, *41*, 12481-12489.
9. Wang, A. Xing, P. Zheng, X. Cao, H. Yang, G. Zheng, X., Deep eutectic solvent catalyzed Friedel-Crafts alkylation of electron-rich arenes with aldehydes. *RSC Adv.* **2015**, *5*, 59022-59026.
10. Maka, H. Spychaj, T. Adamus, J., Lewis acid type deep eutectic solvents as catalysts for epoxy resin crosslinking. *RSC Adv.* **2015**, *5*, 82813-82821.
11. Tran, P. H. Hang, A. H. T., Deep eutectic solvent-catalyzed arylation of benzoxazoles with aromatic aldehydes. *RSC Adv.* **2018**, *8*, 11127-11133.
12. Azizi, N. Edrisi, M. Abbasi, F., Mesoporous silica SBA-15 functionalized with acidic deep eutectic solvent: A highly active heterogeneous *N*-formylation catalyst under solvent-free conditions. *Appl. Organomet. Chem.* **2018**, *32*, e3901.
13. Tavakol, H. Keshavarzipour, F., Preparation of choline chloride-urea deep eutectic solvent-modified magnetic nanoparticles for synthesis of various 2-amino-4H-pyran derivatives in water solution. *Appl. Organomet. Chem.* **2017**, e3811.
14. Polshettiwar, V. Luque, R. Fihri, A. Zhu, H. Bouhrara, M. Bassat, J. M., Magnetically recoverable nanocatalysts. *Chem. Rev.* **2011**, *111*, 3036-3075.
15. Miao, J. Wan, H. Guan, G., Synthesis of immobilized Brønsted acidic ionic liquid on silica gel as heterogeneous catalyst for esterification. *Catal. Commun.* **2011**, *12*, 353-356.
16. Zhang, Q. Su, H. Luo, J. Wei, Y., A magnetic nanoparticle supported dual acidic ionic liquid: a “quasi-homogeneous” catalyst for the one-pot synthesis of benzoxanthenes. *Green Chem.* **2012**, *14*, 201-208.
17. Safari, J. Zarnegar, Z., Immobilized ionic liquid on superparamagnetic nanoparticles as an effective catalyst for the synthesis of tetrasubstituted imidazoles under solvent-free conditions and microwave irradiation. *C. R. Chimie* **2013**, *16*, 920-928.
18. Liang, X., Novel Ionic Liquid Supported on a Magnetic Core and Its Catalytic Activities. *Ind. Eng. Chem. Res.* **2014**, *53*, 17325-17332.
19. Tavakol, H. Keshavarzipour, F., Preparation of choline chloride-urea deep eutectic solvent-modified magnetic nanoparticles for synthesis of various 2-amino-4H-pyran derivatives in water solution. *Appl. Organomet. Chem.* **2017**, *31*, e3811.

20. Huang, Y. Wang, Y. Pan, Q. Wang, Y. Ding, X. Xu, K. Li, N. Wen, Q., Magnetic graphene oxide modified with choline chloride-based deep eutectic solvent for the solid-phase extraction of protein. *Anal. Chim. Acta* **2015**, 877, 90-99.
21. Lamei, N. Ezoddin, M. Ardestani, M. S. Abdi, K., Dispersion of magnetic graphene oxide nanoparticles coated with a deep eutectic solvent using ultrasound assistance for preconcentration of methadone in biological and water samples followed by GC-FID and GC-MS. *Anal. Bioanal. Chem.* **2017**, 409, 6113-6121.
22. Parvatkar, P. T. Parameswaran, P. S. Tilve, S. G., Recent developments in the synthesis of five- and six-membered heterocycles using molecular iodine. *Chem. Eur. J.* **2012**, 18, 5460-5489.
23. Cecchini, M. M. Charnay, C. De Angelis, F. Lamaty, F. Martinez, J. Colacino, E., Poly(ethylene glycol)-based ionic liquids: properties and uses as alternative solvents in organic synthesis and catalysis. *ChemSusChem* **2014**, 7, 45-65.
24. Mohammadi, A. Keshvari, H. Sandaroos, R. Maleki, B. Rouhi, H. Moradi, H. Sepehr, Z. Damavandi, S., A highly efficient and reusable heterogeneous catalyst for the one-pot synthesis of tetrasubstituted imidazoles. *Appl. Catal. A: Gen.* **2012**, 429-430, 73-78.
25. Gawande, M. B. Bonifacio, V. D. Luque, R. Branco, P. S. Varma, R. S., Solvent-free and catalysts-free chemistry: a benign pathway to sustainability. *ChemSusChem* **2014**, 7, 24-44.
26. Gabla, J. J. Mistry, S. R. Maheria, K. C., An efficient green protocol for the synthesis of tetra-substituted imidazoles catalyzed by zeolite BEA: effect of surface acidity and polarity of zeolite. *Catal. Sci. Technol.* **2017**, 7, 5154-5167.
27. Khodairy, A. Ali, A. M. El-Wassimy, M. T., Synthesis of Novel Chromene, Pyridine, Pyrazole, Pyrimidine, and Imidazole Derivatives via one-pot Multicomponent Reaction. *J. Heterocycl. Chem.* **2017**, 54, 3342-3349.
28. Kannan, V. Sreekumar, K., Clay supported titanium catalyst for the solvent free synthesis of tetrasubstituted imidazoles and benzimidazoles. *J. Mol. Catal. A: Chem.* **2013**, 376, 34-39.
29. Maleki, A. Alrezvani, Z. Maleki, S., Design, preparation and characterization of urea-functionalized $\text{Fe}_3\text{O}_4/\text{SiO}_2$ magnetic nanocatalyst and application for the one-pot multicomponent synthesis of substituted imidazole derivatives. *Catal. Commun.* **2015**, 69, 29-33.
30. Shaabani, A. Sepahvand, H. Hooshmand, S. E. Borjian Boroujeni, M., Design, preparation and characterization of $\text{Cu}/\text{GA}/\text{Fe}_3\text{O}_4@\text{SiO}_2$ nanoparticles as a catalyst for the synthesis of benzodiazepines and imidazoles. *Appl. Organomet. Chem.* **2016**, 30, 414-421.
31. Nejatianfar, M. Akhlaghinia, B. Jahanshahi, R., Cu(II) immobilized on guanidinated epibromohydrin-functionalized $\gamma\text{-Fe}_2\text{O}_3@\text{TiO}_2$ ($\gamma\text{-Fe}_2\text{O}_3@\text{TiO}_2\text{-EG-Cu(II)}$): A highly efficient magnetically separable heterogeneous nanocatalyst for one-pot synthesis of highly substituted imidazoles. *Appl.Organomet. Chem.* **2017**, e4095.
32. A. Teimouri and A. N. Chermahini, An efficient and one-pot synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazoles catalyzed via solid acid nano-catalyst, *J. Mol. Catal. A Chem.*, **2011**, 346, 39-45.
33. S. Samai, G. C. Nandi, P. Singh, and M. S. Singh, L -Proline : an efficient catalyst for the one-pot synthesis of 2 , 4 , 5-trisubstituted, *Tetrahedron*, **2009**, 65, 10155–10161.
34. B. Das, J. Kashanna, K. R. Aravind, and P. Jangili, Synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted imidazoles in water using p -dodecylbenzenesulfonic acid as catalyst, *Monatsh Chem.*, **2012**, 44, 223–226.
35. S. E. Wolkenberg, D. D. Wisnoski, W. H. Leister, Y. Wang, Z. Zhao, and C. W. Lindsley, Efficient Synthesis of Imidazoles from Aldehydes and 1 , 2-Diketones Using Microwave Irradiation, *Organic Letters*, **2004**, 6, 1453–1456.