Synthesis of Novel Organosilicon Compounds Possessing Fully Substituted Imidazole Nucleus Sonocatalyzed by Cu-Fe/ZSM-5 Bimetallic Oxides

Kazem D. Safa,^{*a} Aynaz Feyzi,^a Maryam Allahvirdinesbat,^a Leila Sarchami,^a Parvaneh Nakhostin Panahi^b

^aOrganosilicon Research Laboratory, Faculty of Chemistry, University of Tabriz, 5166616471 Tabriz, Iran

^bDepartment of Chemistry, Faculty of Science, University of Zanjan, Iran

Corresponding author. email: dsafa@tabrizu.ac.ir

SUPPLEMENTARY MATERIALS

General procedure for preparation of 1,2,4,5-tetrasubstituted imidazoles

In a 50 mL round bottom flask benzil (1 mmol), aldehyde (1 mmol), amine (1mmol), and ammonium acetate (1.1 mmol) were taken in the presence of 3wt.% of Fe-Cu/ZSM-5 in 10 mL of ethanol under ultrasonic irradiation. After completion of the reaction (monitored by TLC, within 2-5 min), the crude residue was cooled to room temperature. Then catalyst was separated by filtration. The filtrate was evaporated to give the crude product. This product was washed with n-hexane and purified by recrystallization from ethanol. The supported reagent was washed thrice with ethanol and dried under vacuum before reuse.

Spectral data of some representative products are given below:

4-(2,4,5-triphenyl-1H-imidazol-1-yl)phenol (1a)

White powder (99%), M.P. >280 °C, IR (KBr, cm^{-1}): ν_{max} = 3414, 3054, 2922, 1604, 1513, 1446, 696. ^1H NMR (DMSO- d_6 , 400 MHz) δ_{H} : 6.66 (d, J = 8.5 Hz, 2H, Ar-H), 7.04 (d, J = 8.5 Hz, 2H, Ar-H), 7.14-7.30 (m, 11H, Ar-H), 7.42-7.44 (m, 2H, Ar-H), 7.49 (d, J 7.5, 2H, Ar-H), 9.78 (s, 1H, Ar-OH). (DMSO- d_6 , 100 MHz) δ_{C} : 115.59, 126.35, 127.84, 128.12, 128.15, 128.27, 128.40, 129.79, 130.58, 130.63, 131.12, 131.59, 134.56, 136.61, 146.11, 157.31. Anal.Calc. for $\text{C}_{27}\text{H}_{20}\text{N}_2\text{O}$: C, 83.48; H, 5.19; N, 7.21%. Found: C, 83.54; H, 5.07; N, 7.19%.

^1H NMR spectrum of 4-(2,4,5-triphenyl-1H-imidazol-1-yl)phenol (**1a**) in DMSO- d_6

^{13}C NMR spectrum of 4-(2,4,5-triphenyl-1H-imidazol-1-yl)phenol (**1a**) in DMSO-d_6

4-(2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazol-1-yl)phenol (1c)

White powder (99%), M.P. >280 °C, IR (KBr, cm^{-1}): ν_{max} = 3416, 3054, 2926, 1595, 1513, 1478, 1440, 745, 695.

^1H NMR (DMSO- d_6 , 400 MHz) δ_{H} : 6.67 (d, J = 8.4 Hz, 2H, Ar-H), 7.05 (d, J = 8.4 Hz, 2H, Ar-H), 7.14-7.43 (m, 12H, Ar-H), 7.47 (d, J = 7.5 Hz, 2H, Ar-H), 9.82 (s, 1H, Ar-OH). (DMSO- d_6 , 100 MHz) δ_{C} : 115.69, 126.33, 126.46, 127.55, 128.15, 128.27, 128.37, 128.43, 129.37, 129.71, 129.74, 130.44, 131.10, 131.89, 132.98, 134.37, 136.76, 144.96, 157.42. Anal.Calc. for $\text{C}_{27}\text{H}_{19}\text{ClN}_2\text{O}$: C, 76.68; H, 4.53; N, 6.62%. Found: C, 76.49; H, 4.61; N, 6.58%.

^1H NMR spectrum of 4-(2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazol-1-yl)phenol (**1c**) in DMSO- d_6

¹³C NMR spectrum of 4-(2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazol-1-yl)phenol (**1c**) in DMSO-d₆

4,4'-(4,5-diphenyl-1*H*-imidazole-1,2-diyl)diphenol (1d)

White powder (97%), M.P. >280 °C, IR (KBr, cm^{-1}): ν_{max} = 3416, 3053, 2924, 1642, 1513, 1438, 694. ^1H NMR (DMSO- d_6 , 400 MHz) δ_{H} : 6.64-6.67 (m, 4H, Ar-H), 7.00 (d, J = 8.4, 2H, Ar-H), 7.12-7.29 (m, 10H, Ar-H), 7.45 (d, J = 7.7, 2H, Ar-H), 9.73 (bs, 2H, Ar-OH). (DMSO- d_6 , 100 MHz) δ_{C} : 114.92, 115.55, 121.48, 126.22, 126.31, 128.05, 128.09, 128.16, 128.39, 129.67, 129.84, 130.86, 130.89, 131.14, 134.75, 136.15, 146.56, 157.19, 157.52. Anal.Calc. for $\text{C}_{27}\text{H}_{20}\text{N}_2\text{O}_2$: C, 80.18; H, 4.98; N, 6.93%. Found: C, 80.01; H, 4.75; N, 6.87%.

¹H NMR spectrum of 4,4'-(4,5-diphenyl-1H-imidazole-1,2-diyl)diphenol (**1d**) in DMSO-d₆

¹³C NMR spectrum of 4,4'-(4,5-diphenyl-1H-imidazole-1,2-diyl)diphenol (**1d**) in DMSO-d₆

2-(4-Chlorophenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (1f)

White powder (97%), M.P. = 167-169 °C, IR (KBr, cm^{-1}): ν_{max} = 3054, 2976, 1510, 1478, 1445, 745, 696. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.36 (s, 3H, CH_3), 6.95 (d, J = 8.1 Hz, 2H, Ar-H), 7.10 (d, J = 8.1 Hz, 2H, Ar-H), 7.16-7.18 (m, 2H, Ar-H), 7.21-7.30 (m, 8H, Ar-H), 7.41-7.43 (m, 2H, Ar-H), 7.61-7.63 (m, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.14 (CH_3), 119.77, 125.61, 126.30, 126.95, 126.99, 127.13, 127.29, 127.31, 128.10, 128.82, 129.03, 129.50, 130.05, 130.17, 133.15, 133.21, 133.31, 137.41, 144.74. Anal.Calc. for $\text{C}_{28}\text{H}_{21}\text{ClN}_2$: C, 79.89; H, 5.03; N, 6.66%. *Found*: C, 79.76; H, 5.14; N, 6.86%.

¹H NMR spectrum of 2-(4-Chlorophenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**1f**) in CDCl₃

¹³C NMR spectrum of 2-(4-chlorophenyl)-4,5-diphenyl-1-p-tolyl-1H-imidazole (**1f**) in CDCl₃

1-(4-Bromophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole 1g

White powder (98%), M.P. = 190-192 °C, IR (KBr, cm^{-1}): ν_{max} = 3445, 3057, 2961, 2908, 1570, 1510, 1478, 726, 697. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.36 (s, 3H, CH_3), 6.93 (d, J = 8.6 Hz, 2H, Ar-H), 7.11-7.17 (m, 4H, Ar-H), 7.20-7.35 (m, 8H, Ar-H), 7.41 (d, 2H, J = 8.6 Hz, Ar-H), 7.60-7.62 (m, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.27 (CH_3), 121.06, 121.58, 125.64, 126.37, 127.14, 127.51, 127.88, 127.96, 128.56, 128.91, 129.35, 129.44, 130.10, 131.13, 131.25, 133.27, 135.27, 137.42, 146.04. Anal.Calc. for $\text{C}_{28}\text{H}_{21}\text{BrN}_2$: C, 72.26; H, 4.55; N, 6.02%.
Found: C, 72.14; H, 4.47; N, 6.14%.

¹H NMR spectrum of 1-(4-Bromophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**1g**) in CDCl₃

¹³C NMR spectrum of 1-(4-Bromophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**1g**) in CDCl₃

1-(3-Chlorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (1i)

White powder (99%), M.P. = 198-200 °C, IR (KBr, cm^{-1}): ν_{max} = 3056, 2963, 2909, 1490, 1479, 739, 703. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.33 (s, 3H, CH_3), 6.93-6.95 (m, 1H, Ar-H), 7.05 (t, J = 1.86 Hz, 1H, ArH), 7.08 (d, 2H, J = 8.04 Hz, Ar-H), 7.12-7.27 (m, 10H, Ar-H), 7.32 (d, 2H, J = 8.15 Hz, Ar-H), 7.58-7.60 (m, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.27 (CH_3), 125.64, 125.77, 126.24, 126.33, 127.14, 127.15, 127.47, 127.58, 127.78, 127.95, 128.92, 129.31, 130.05, 133.21, 133.48, 137.33, 137.45, 146.00. Anal.Calc. for $\text{C}_{28}\text{H}_{21}\text{ClN}_2$: C, 79.89; H, 5.03; N, 6.66%. *Found*: C, 79.78; H, 5.12; N, 6.74%.

¹H NMR spectrum of 1-(3-Chlorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**1i**) in CDCl₃

¹³C NMR spectrum of 1-(3-Chlorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**1i**) in CDCl₃

1-(4-Fluorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (11)

White powder (97%), M.P. = 188-190 °C, IR (KBr, cm^{-1}): ν_{max} = 3058, 2962, 2908, 1602, 1508, 1444, 960, 775, 730. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.36 (s, 3H, CH_3), 6.96-7.00 (m, 2H, Ar-H), 7.03-7.07 (m, 2H, Ar-H), 7.11 (d, 2H, J = 8.0 Hz, Ar-H), 7.14-7.17 (m, 2H, Ar-H), 7.21-7.32 (m, 6H, Ar-H), 7.35 (d, 2H, J = 8.0 Hz, Ar-H), 7.63 (d, 2H, J = 7.2 Hz, Ar-H), ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.24 (CH_3), 114.95, 115.17, 125.58, 126.32, 126.41, 127.01, 127.12, 127.41, 127.79, 127.88, 128.99, 129.08, 129.50, 129.58, 130.07, 132.19, 133.30, 137.16, 137.31, 146.12, 159.59, 162.06. Anal.Calc. for $\text{C}_{28}\text{H}_{21}\text{FN}_2$: C, 83.14; H, 5.23; N, 6.93%. *Found*: C, 83.09; H, 5.14; N, 6.81%.

¹H NMR spectrum of 1-(4-Fluorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**II**) in CDCl₃

¹³C NMR spectrum of 1-(4-Fluorophenyl)-4,5-diphenyl-2-*p*-tolyl-1*H*-imidazole (**11**) in CDCl₃

General procedure for bromination of imidazole derivatives

A mixture of (1 mmol) imidazole derivative (**1a** or **1c**) and (1 mmol) dibromoalkane (1,2-dibromoethane, 1,3-dibromopropane or 1,4-dibromobutane), also (1 mmol) imidazole derivative (**1d**) and (1 mmol) 1,4-dibromobutane in presence of K_2CO_3 in 100 ml of dry DMF was stirred at room temperature for 24 h. Then DMF solvent was evaporated in vacuum, and the residue was taken up in CH_2Cl_2 (50 ml) and the extract was washed with brine (30 ml) and then dried (Na_2SO_4). After filtration the solvent evaporated, and the residue was purified by silica gel column chromatography using hexane/ethylacetate (75:25, v/v). Fraction containing 4-bromoalkoxy-imidazole was collected then the solvent was removed under reduced pressure.

Spectral data of some novel representative products are given below:

1-(4-(4-bromobutoxy)phenyl)-2,4,5-triphenyl-1*H*-imidazole (**2a**)

Yellow powder (69%), M.P. = 132-134 °C, IR (KBr, cm^{-1}): ν_{max} = 3058, 2924, 1609, 1510, 1469, 1165, 1101, 695. 1H NMR ($CDCl_3$, 400 MHz): δ_H 1.90-1.95 (m, 2H, CH_2), 2.01-2.08 (m, 2H, CH_2), 3.48 (t, J 6.5, 2H, CH_2), 3.94 (t, J 6.0, 2H, CH_2), 6.73 (d, J 8.8, 2H, Ar-H), 6.94 (d, J 8.8, 2H, Ar-H), 7.12-7.15 (m, 2H, Ar-H), 7.16-7.26 (m, 9H, Ar-H), 7.43-7.46 (m, 2H, Ar-H), 7.58 (d, J = 7.3, 2H, Ar-H). ^{13}C NMR ($CDCl_3$, 100 MHz): δ_C 28.3, 28.67, 32.30, 65.96 (CH_2), 113.61, 125.49, 126.34, 126.85, 127.07, 127.09, 127.31, 127.89, 127.88, 128.15, 128.42, 128.90, 129.62, 129.76, 130.12, 133.50, 146.00, 157.37. Anal.Calc. for $C_{31}H_{27}BrN_2O$: C, 71.13; H, 5.20; N, 5.35%. Found: C, 71.33; H, 5.38; N, 5.21%.

^1H NMR spectrum of 1-(4-(4-bromobutoxy)phenyl)-2,4,5-triphenyl-1H-imidazole (**2a**) in CDCl₃

¹³C NMR spectrum of 1-(4-(4-bromobutoxy)phenyl)-2,4,5-triphenyl-1H-imidazole (**2a**) in CDCl₃

2-(4-(2-bromoethoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (2b)

White powder (60%), M.P. = 164-166 °C, IR (KBr, cm^{-1}) = 3057, 2924, 1617, 1539, 1512, 1457, 1130, 615. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.32 (s, 3H, CH_3), 3.62 (t, $J = 6.3$ Hz, 2H, CH_2), 4.26 (t, $J = 6.3$ Hz, 2H, CH_2), 6.78 (d, $J = 8.8$ Hz, 2H, Ar-H), 6.91 (d, $J = 8.2$ Hz, 2H, Ar-H), 7.05 (d, $J = 8.2$ Hz, 2H, Ar-H), 7.11-7.26 (m, 8H, Ar-H), 7.37 (d, $J = 7.0$ Hz, 2H, Ar-H), 7.58 (d, $J = 7.2$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.16 (CH_3), 27.94, 66.68 (CH_2), 113.24, 123.02, 125.47, 126.36, 126.80, 127.11, 127.12, 127.27, 128.69, 129.37, 129.61, 129.80, 130.12, 133.52, 137.11, 145.68, 156.96. Anal.Calc. for $\text{C}_{30}\text{H}_{25}\text{BrN}_2\text{O}$: C, 70.73; H, 4.95; N, 5.50%. *Found*: C, 70.63; H, 4.86; N, 5.41%.

¹H NMR spectrum of 2-(4-(2-bromoethoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**2b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(2-bromoethoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**2b**) in CDCl₃

1-(4-(4-bromobutoxy) phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1*H*-imidazole (2c)

White powder (64%), M.P. = 148-150 °C, IR (KBr, cm^{-1}) = 3057, 2927, 1650, 1605, 1562, 1541, 1511, 1475, 1417, 1244, 1167, 1091, 1014, 777, 754, 725, 695, 657, 542. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 1.90-1.96 (m, 2H, CH_2), 2.02-2.09 (m, 2H, CH_2), 3.48 (t, J = 6.5 Hz, 2H, CH_2), 3.94 (t, J = 6.0 Hz, 2H, CH_2), 6.72-6.76 (m, 2H, Ar-H), 6.91-6.95 (m, 2H, Ar-H), 7.11-7.14 (m, 2H, Ar-H), 7.16-7.26 (m, 8H, Ar-H), 7.36-7.39 (m, 2H, Ar-H), 7.55-7.57 (m, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 26.74, 28.35, 32.37, 65.97 (CH_2), 113.73, 125.54, 125.63, 126.29, 126.97, 127.16, 127.34, 127.36, 128.05, 128.34, 128.57, 129.01, 129.47, 130.05, 130.30, 133.16, 133.28, 137.24, 144.84, 157.49. Anal.Calc. for $\text{C}_{31}\text{H}_{26}\text{BrClN}_2\text{O}$: C, 66.74; H, 4.70; N, 5.02%. *Found*: C, 66.63; H, 4.88; N, 5.31%.

¹H NMR spectrum of 1-(4-(4-bromobutoxy) phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**2c**) in CDCl₃

¹³C NMR spectrum of 1-(4-(4-bromobutoxy) phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**2c**) in CDCl₃

1,2-bis(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1H-imidazole (2d)

White powder (59%), M.P. = 171-173°C, IR (KBr, cm^{-1}) = 3057, 2924, 1616, 1539, 1512, 1462, 1109, 1028, 695.

^1H NMR (CDCl_3 , 400 MHz): δ_{H} 1.95-1.98 (m, 4H, CH_2), 2.06-2.11 (m, 4H, CH_2), 3.52 (t, $J = 6.3$ Hz, 4H, CH_2), 3.97-4.02 (m, 4H, CH_2), 6.78-6.81 (m, 4H, Ar-H), 6.98 (d, $J = 8.7$ Hz, 2H, Ar-H), 7.16-7.30 (m, 8H, Ar-H), 7.40 (d, $J = 8.7$ Hz, 2H, Ar-H), 7.61 (d, $J = 7.3$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 26.79, 28.40, 28.43, 32.33, 32.42, 65.76, 66.00 (CH_2), 113.05, 113.65, 125.50, 126.39, 126.82, 127.11, 127.31, 128.48, 130.15, 133.55, 133.58, 136.91, 157.77, 157.97. Anal.Calc. for $\text{C}_{35}\text{H}_{34}\text{Br}_2\text{N}_2\text{O}_2$: C, 62.33; H, 5.08; N, 4.15%. *Found*: C, 62.45; H, 4.98; N, 4.31%.

¹H NMR spectrum of 1,2-bis(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1H-imidazole (**2d**) in CDCl₃

¹³C NMR spectrum of 1,2-bis(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1H-imidazole (**2d**) in CDCl₃

2-(4-(3-bromopropoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1H-imidazole (3b)

White powder (60%), M.P. = 160-162 °C, IR (KBr, cm^{-1}) = 3056, 2924, 1605, 1577, 1513, 1465, 1245, 1175, 1108, 1026, 698. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 2.30-2.35 (m, 5H, CH_2 and CH_3), 3.62 (t, $J = 6.4$ Hz, 2H, CH_2), 4.11 (t, $J = 5.8$ Hz, 2H, CH_2), 6.81 (d, $J = 8.6$ Hz, 2H, Ar-H), 6.95 (d, $J = 8.0$ Hz, 2H, Ar-H), 7.09 (d, $J = 7.9$ Hz, 2H, Ar-H), 7.15-7.30 (m, 8H, Ar-H), 7.40 (d, $J = 8.6$ Hz, 2H, Ar-H), 7.62 (d, $J = 7.3$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.15 (CH_3), 28.93, 31.30, 64.20 (CH_2), 113.05, 122.46, 125.46, 126.37, 126.53, 126.78, 127.09, 127.13, 127.26, 128.19, 128.67, 129.30, 129.55, 129.82, 130.12, 133.54, 136.90, 137.07, 145.82, 157.61. Anal.Calcd for $\text{C}_{31}\text{H}_{27}\text{BrN}_2\text{O}$: C, 71.13; H, 5.20; N, 5.35%. Found: C, 70.98; H, 5.04; N, 5.24%.

¹H NMR spectrum of 2-(4-(3-bromopropoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**3b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(3-bromopropoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**3b**) in CDCl₃

2-(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (4b)

White powder (62%), M.P. = 182-184 °C, IR (KBr, cm^{-1}) = 3058, 2927, 1639, 1616, 1513, 1469, 1241, 1015, 829, 776, 699. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 1.90-1.95 (m, 2H, CH_2), 2.01-2.08 (m, 2H, CH_2), 2.31 (s, 3H, CH_3), 3.48 (t, $J = 6.6$ Hz, 2H, CH_2), 3.96 (t, $J = 6.0$ Hz, 2H, CH_2), 6.75 (d, $J = 8.8$ Hz, 2H, Ar-H), 6.91 (d, $J = 8.1$ Hz, 2H, Ar-H), 7.04 (d, $J = 8.1$ Hz, 2H, Ar-H), 7.11-7.26 (m, 8H, Ar-H), 7.35 (d, $J = 8.8$ Hz, 2H, ArH), 7.58 (d, $J = 7.3$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 20.17 (CH_3), 26.79, 28.40, 32.47, 65.69 (CH_2), 112.95, 125.44, 126.37, 126.76, 127.11, 127.26, 128.66, 129.28, 129.49, 129.80, 130.11, 133.53, 137.03, 145.86, 157.75. Anal.Calc. for $\text{C}_{32}\text{H}_{29}\text{BrN}_2\text{O}$: C, 71.51; H, 5.44; N, 5.21%. *Found*: C, 71.39; H, 5.51; N, 5.24%.

¹H NMR spectrum of 2-(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**4b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(4-bromobutoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**4b**) in CDCl₃

General procedure for preparation of imidazoles containing bulky organosilicon groups

To a stirred solution of $(\text{Me}_3\text{Si})_3\text{CLi}$ (5 mmol) in THF was added a bromo-imidazole derivative (5 mmol) in 10 ml THF and or (2.5 mmol) of **2d** compound in THF at $^\circ\text{C}$. The mixture was stirred for another 10 min at room temperature. The mixture was poured into ammonium chloride solution (50 ml) and extracted with CH_2Cl_2 (2×20 ml). The organic phase was washed with water (100 mL) and dried (Na_2SO_4), and the solvent was removed in vacuum to yield a yellow solid.

Selected spectral data of the products:

1-(4-(5,5,5-tris(trimethylsilyl)pentyl)oxy)phenyl-2,4,5-triphenyl-1H-imidazole (**3a**)

White powder (90%), M.P. = 164-166 $^\circ\text{C}$, IR (KBr, cm^{-1}) = 3061, 2953, 1611, 1510, 1471, 1445, 1255, 1029, 840, 695, 673, 641, 615, 536. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.15 (s, 27H, SiMe_3), 1.70-1.79 (m, 6H, $3 \times \text{CH}_2$), 3.93 (t, J = 6.0 Hz, 2H, CH_2), 6.77 (d, J = 8.8 Hz, 2H, Ar-H), 6.98 (d, J = 8.8 Hz, 2H, Ar-H), 7.16-7.36 (m, 11H, Ar-H), 7.47-7.50 (m, 2H, Ar-H), 7.62 (d, J = 7.1 Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.75 (SiMe_3), 5.14 ($\text{C}(\text{SiMe}_3)_3$), 25.46, 29.52, 29.83, 66.60 (CH_2), 113.67, 125.50, 126.38, 126.84, 127.07, 127.11, 127.31, 127.90, 128.10, 128.38, 128.70, 129.66, 129.79, 130.14, 133.55, 137.09, 157.57. Anal.Calc. for $\text{C}_{41}\text{H}_{54}\text{N}_2\text{OSi}_3$: C, 72.94; H, 8.06; N, 4.15. Found: C, 72.79; H, 8.11; N, 3.94%.

¹H NMR spectrum of 1-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-2,4,5-triphenyl-1H-imidazole (**3a**) in CDCl₃

¹³C NMR spectrum of 1-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-2,4,5-triphenyl-1H-imidazole (**3a**) in CDCl₃

1-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1*H*-imidazole (3c)

White powder (91%), M.P. = 168-170 °C, IR (KBr, cm^{-1}) = 3059, 2958, 1618, 1541, 1512, 1473, 1256, 1094, 840, 620. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.10 (s, 27H, SiMe_3), 1.62-1.66 (m, 6H, $3\times\text{CH}_2$), 3.85 (t, $J = 5.9$ Hz, 2H, CH_2), 6.69 (d, $J = 8.8$ Hz, 2H, Ar-H), 6.88 (d, $J = 8.8$ Hz, 2H, Ar-H), 7.06 (dd, $J = 1.7$ Hz, $J = 7.42$ Hz, 2H, Ar-H), 7.11-7.23 (m, 8H, Ar-H), 7.33 (d, $J = 8.5$ Hz, 2H, Ar-H), 7.50 (d, $J = 7.2$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.72 (SiMe_3), 4.43 ($\text{C}(\text{SiMe}_3)_3$), 25.46, 29.51, 29.83, 66.60 (CH_2), 113.95, 126.18, 126.37, 126.59, 127.28, 127.50, 127.53, 128.02, 128.24, 128.42, 128.81, 128.87, 129.38, 129.68, 130.09, 130.12, 133.00, 137.04, 157.31. Anal.Calc. for $\text{C}_{41}\text{H}_{53}\text{ClN}_2\text{OSi}_3$: C, 69.40; H, 7.53; N, 3.95%. *Found*: C, 69.48; H, 7.32; N, 3.79%.

¹H NMR spectrum of 1-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**3c**) in CDCl₃

¹³C NMR spectrum of 1-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole (**3c**) in CDCl₃

1,2-bis(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-4,5-diphenyl-1*H*-imidazole (3d)

White powder (89%), M.P. = 200-202 °C, IR (KBr, cm^{-1}) = 3058, 2925, 1617, 1558, 1540, 1510, 1457, 1250, 841, 619. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.05 (s, 27H, SiMe_3), 0.06 (s, 27H, SiMe_3), 1.54-1.63 (m, 12H, CH_2), 3.82-3.87 (m, 4H, CH_2), 6.66-6.72 (m, 4H, Ar-H), 6.88 (d, J = 8.8 Hz, 2H, Ar-H), 7.01-7.21 (m, 8H, Ar-H), 7.30 (d, J = 8.8, 2H, Ar-H), 7.52 (d, J = 7.1, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.75 (SiMe_3), 4.43 ($\text{C}(\text{SiMe}_3)_3$), 25.51, 28.69, 66.84, 65.99 (CH_2), 113.05, 113.63, 125.49, 126.38, 126.82, 127.09, 127.27, 128.42, 129.21, 130.14, 133.52, 133.56, 136.83, 157.77, 157.97. Anal.Calc. for $\text{C}_{55}\text{H}_{88}\text{N}_2\text{O}_2\text{Si}_6$: C, 67.56; H, 9.07; N, 2.86%. Found: C, 67.44; H, 9.28; N, 2.65%.

¹H NMR spectrum of 1,2-bis(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-4,5-diphenyl-1H-imidazole (**3d**) in CDCl₃

¹³C NMR spectrum of 1,2-bis(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-4,5-diphenyl-1H-imidazole (**3d**) in CDCl₃

2-(4-(3,3,3-tris(trimethylsilyl)propoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (5b)

White powder (89%), M.P. = 106-108°C, IR (KBr, cm^{-1}) = 3058, 2955, 1609, 1578, 1512, 1475, 1422, 1256, 1024, 845, 696. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.18 (s, 27H, SiMe_3), 2.15 (t, $J = 8.2$ Hz, 2H, CH_2), 2.34 (s, 3H, CH_3), 4.08 (t, $J = 8.2$ Hz, 2H, CH_2), 6.74 (d, $J = 8.8$ Hz, 2H, Ar-H), 6.93 (d, $J = 8.2$ Hz, 2H, Ar-H), 7.07 (d, $J = 8.2$ Hz, 2H, Ar-H), 7.13-7.28 (m, 8H, Ar-H), 7.37 (d, $J = 7.9$ Hz, 2H, Ar-H), 7.60 (d, $J = 7.9$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.72 (SiMe_3), 4.40 ($\text{C}(\text{SiMe}_3)_3$), 20.18 (CH_3), 29.10, 67.49 (CH_2), 113.06, 125.42, 126.36, 126.76, 127.09, 127.12, 127.26, 128.67, 129.33, 129.89, 130.14, 133.63, 137.02, 146.00, 158.00. Anal.Calc. for $\text{C}_{40}\text{H}_{52}\text{N}_2\text{OSi}_3$: C, 72.67; H, 7.93; N, 4.24%. *Found*: C, 72.46; H, 7.85; N, 4.11%.

¹H NMR spectrum of 2-(4-(3,3,3-tris(trimethylsilyl)propoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**5b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(3,3,3-tris(trimethylsilyl)propoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**5b**) in CDCl₃

2-(4-(4,4,4-tris(trimethylsilyl)butoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (6b)

White powder (93%), M.P. = 140-142 °C, IR (KBr, cm^{-1}) = 3059, 2954, 1616, 1511, 1472, 1253, 1176, 1027, 898, 836, 698. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.13 (s, 27H, SiMe_3), 1.76 (t, J = 3.9 Hz, 2H, CH_2), 1.93-2.01 (m, 2H, CH_2), 2.31 (s, 3H, CH_3), 3.87 (t, J = 6.3 Hz, 2H, CH_2), 6.76 (d, J = 8.7 Hz, 2H, Ar-H), 6.91 (d, J = 8.2 Hz, 2H, Ar-H), 7.04 (d, J = 8.2 Hz, 2H, Ar-H), 7.11-7.30 (m, 8H, Ar-H), 7.35 (d, J = 8.7 Hz, 2H, Ar-H), 7.58 (d, J = 7.3 Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.72 (SiMe_3), 4.43 ($\text{C}(\text{SiMe}_3)_3$), 20.17 (CH_3), 26.10, 28.70, 67.56 (CH_2), 113.02, 125.42, 126.39, 126.74, 127.09, 127.14, 127.25, 128.65, 129.25, 129.86, 130.12, 133.59, 136.99, 145.98, 157.98. Anal.Calc. for $\text{C}_{41}\text{H}_{54}\text{N}_2\text{OSi}_3$: C, 72.94; H, 8.06; N, 4.15%. *Found*: C, 72.68; H, 7.94; N, 4.21%.

¹H NMR spectrum of 2-(4-(4,4,4-tris(trimethylsilyl)butoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**6b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(4,4,4-tris(trimethylsilyl)butoxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1H-imidazole (**6b**) in CDCl₃

2-(4-(5,5,5-tris(trimethylsilyl)pentyl)oxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (7b)

White powder (92%), M.P. = 202-204 °C, IR (KBr, cm^{-1}) = 3057, 2951, 1608, 1577, 1514, 1476, 1254, 1174, 1030, 840, 696. ^1H NMR (CDCl_3 , 400 MHz): δ_{H} 0.17 (s, 27H, SiMe_3), 1.72-1.78 (m, 6H, CH_2), 2.36 (s, 3H, CH_3), 3.97 (t, $J = 5.9$ Hz, 2H, CH_2), 6.80 (d, $J = 8.9$ Hz, 2H, Ar-H), 6.96 (d, $J = 8.2$ Hz, 2H, Ar-H), 7.09 (d, $J = 8.1$ Hz, 2H, Ar-H), 7.16-7.31 (m, 8H, Ar-H), 7.40 (d, $J = 8.8$ Hz, 2H, Ar-H), 7.63 (d, $J = 7.1$ Hz, 2H, Ar-H). ^{13}C NMR (CDCl_3 , 100 MHz): δ_{C} 1.72 (SiMe_3), 4.38 ($\text{C}(\text{SiMe}_3)_3$), 20.16 (CH_3), 25.50, 29.60, 29.83, 66.29 (CH_2), 112.99, 121.99, 125.39, 126.35, 126.71, 127.06, 127.10, 127.23, 128.62, 129.21, 129.84, 130.09, 133.55, 133.58, 136.94, 157.92. Anal.Calc. for $\text{C}_{42}\text{H}_{56}\text{N}_2\text{OSi}_3$: C, 73.20; H, 8.19; N, 4.06%. *Found*: C, 73.09; H, 8.23; N, 4.11%.

¹H NMR spectrum of 2-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**7b**) in CDCl₃

¹³C NMR spectrum of 2-(4-(5,5,5-tris(trimethylsilyl)pentyloxy)phenyl)-4,5-diphenyl-1-*p*-tolyl-1*H*-imidazole (**7b**) in CDCl₃

S1. SEM image of H-ZSM-5

S2. TEM image of H-ZSM-5

S3. SEM image of Cu/ZSM-5

S4. TEM image of Cu /ZSM-5

S5. SEM image of Cu-Fe/ZSM-5

S6. TEM image of Fe-Cu/ZSM-5

S7. The XRD patterns of Fe-Cu/ZSM-5