Supplementary Material (ESI) for Chemical Communications

Supplementary information

Iodine-mediated cyclization of *N*-thioacyl 1-(2-pyridyl)-1,2-aminoalcohols and their subsequent condensation leading to formation of novel bis(1-imidazo[1,5-a]pyridyl)arylmethanes

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Bis(3-pheny-1-limidazo[1,5-a]pyridyl)phenylmethane (**3a**) (ST287)

To a THF solution (2 mL) of *N*-(2-hydroxy-2-phenyl-1-(2-pyridyl)ethyl)benzenecarbo-thioamide (**1a**) (227 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-phenyl-1-imidazo[1,5-a]pyridyl)phenylmethane (**3a**) (0.111 g, 0.023 mmol, 93% per 0.25 mmol) as a green solid; mp 103-104 °C; IR (KBr) 3372, 3207, 2709, 2318, 1961, 1894, 1814, 1748, 1713, 1660, 1597, 1455, 1366, 1184, 1124, 1066, 1027, 970, 918, 882, 842, 795, 694, 613, 577, 521, 480 cm^{-1}; ^1H NMR ($CDCl_3$) δ 6.26 (s, 1H, C<u>H</u>-Ar_3), 6.29 (dd, *J* = 7.3, 6.3 Hz, 2H, Ar), 6.40 (dd, *J* = 9.3, 6.3 Hz, 2H, Ar), 7.07 (t, *J* = 7.3 Hz, 1H, Ar), 7.16 (t, *J* = 7.8 Hz, 2H, Ar), 7.23 (t, *J* = 7.3 Hz, 2H, Ar), 7.33 (t, *J* = 7.8 Hz, 4H, Ar), 7.36 (d, *J* = 9.3 Hz, 2H, Ar), 7.39 (d, *J* = 7.3 Hz, 2H, Ar), 7.63 (d, *J* = 7.8 Hz, 4H, Ar), 8.01 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR δ 44.1 (<u>C</u>H-Ar_3), 112.8, 117.6, 119.6, 121.0, 126.1, 128.0, 128.1, 128.2, 128.5, 128.7, 128.8, 130.5, 133.9, 136.6, 143.1 (Ar); MS (El) *m/z* 476 (M^+); HRMS calcd for $C_{33}H_{24}N_4$: 476.1995, found: 476.1984.

Bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3b**) (ST291)

To a THF solution (2 mL) of *N*-[2-hydroxyl-2-(4-methoxyphenyl)-1-(2-pyridyl)propyl]benzenecarbothioamide (**1b**) (182 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3b**) (0.111 g, 0.018 mmol, 72% per 0.25 mmol) as a green solid; mp 95–96 °C; IR (KBr) 3302, 3063, 2927, 2834, 1748, 1664, 1602, 1510, 1458, 1364, 1300, 1247, 1172, 1077, 1029, 960, 910, 885, 793, 774, 729, 692, 614, 588 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.67 (s, 3H, -OC<u>H</u>$_3$), 6.23 (s, 1H, C<u>H</u>-Ar_3), 6.36 (ddd, *J* = 7.3, 6.3, 1.0 Hz, 2H, Ar), 6.45 (ddd, *J* = 9.3, 6.3, 1.0 Hz, 2H, Ar), 6.74 (d, *J* = 8.8 Hz, 2H, Ar), 7.28 (t, *J* = 7.3 Hz, 2H, Ar), 7.33 (d, *J* = 8.8 Hz, 2H, Ar), 7.36 (d, *J* = 9.3 Hz, 2H, Ar), 7.38 (t, *J*

= 7.3 Hz, 4H, Ar), 7.68 (dd, *J* = 7.3, 1.5 Hz, 4H, Ar), 8.07 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR δ 44.1 (CH-Ar$_3$), 55.1 (CH$_3$O-), 112.9, 113.6, 117.7, 119.7, 121.1, 128.2, 128.3, 128.4 128.9, 129.8, 130.5, 134.2, 135.3, 136.6, 158.0 (Ar); MS (El) *m/z* 506 (M^+); HRMS calcd for $C_{34}H_{26}N_4O$: 506.2107, found: 506.2107.

Bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3c**) (ST298)

To a THF solution (4 mL) of *N*-(2-(4-chlorophenyl)-2-hydroxy-1-(2-pyridyl)ethyl)benzenecarbothioamide (**1c**) (184 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3c**) (0.104 g, 0.020 mmol, 81% per 0.25 mmol) as a green solid; mp 77-78 °C; IR (KBr) 3063, 1900, 1749, 1711, 1667, 1602, 1488, 1221, 1177, 1089, 1015, 962, 918, 883, 773, 732, 697, 615, 531, 499, 449 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 6.22 (s, 1H, CH-Ar$_3$), 6.34 (dd, *J* = 6.8, 6.3 Hz, 2H, Ar), 6.45 (dd, *J* = 9.3, 6.3 Hz, 2H, Ar), 7.14 (d, *J* = 8.3 Hz, 2H, Ar), 7.26 (t, *J* = 7.3 Hz, 2H, Ar), 7.33 (d, *J* = 8.3 Hz, 2H, Ar), 7.36 (t, *J* = 7.3 Hz, 4H, Ar), 7.42 (d, *J* = 9.3 Hz, 2H, Ar), 7.65 (d, *J* = 7.3 Hz, 4H, Ar), 8.05 (d, *J* = 6.8 Hz, 2H, Ar); ^{13}C NMR δ 44.1 (CH-Ar$_3$), 113.0, 118.0, 119.4, 121.1, 128.1, 128.2, 128.4, 128.5, 128.9, 130.1, 130.4, 131.9, 133.3, 136.7, 141.7 (Ar); MS (El) *m/z* 510 (M^+); HRMS calcd for $C_{33}H_{23}ClN_4$: 510.1611; found, 510.1632.

Bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-methylphenylmethane (**3d**) (ST299)

To a THF solution (4 mL) of *N*-(2-hydroxyl-1-(2-pyridyl)-2-(4-methylphenyl)ethyl)benzenecarbothioamide (**1d**) (174 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-phenyl-1-imidazo[1,5-a]pyridyl)-4-methylphenylmethane (**3d**) (0.126 g, 0.025 mmol, 100% per 0.25 mmol) as a green solid; mp 116-117 °C; IR (KBr) 3053, 2917, 1903, 1750, 1662,

1631, 1602, 1509, 1458, 1403, 1365, 1315, 1261, 1181, 1127, 1075, 1026, 999, 960, 915, 853, 745, 698, 853, 745, 698, 500, 429 cm^{-1}; ^1H NMR ($CDCl_3$) δ 2.18 (s, 3H, C<u>H</u>$_3$), 6.22 (s, 1H, C<u>H</u>-Ar$_3$), 6.28 (dd, *J* = 7.3, 6.3 Hz, 2H, Ar), 6.39 (dd, *J* = 9.3, 6.3 Hz, 2H, Ar), 6.97 (d, *J* = 7.8 Hz, 2H, Ar), 7.22 (t, *J* = 7.3 Hz, 2H, Ar), 7.30 (t, *J* = 7.3 Hz, 4H, Ar), 7.33 (d, *J* = 7.8 Hz, 2H, Ar), 7.38 (d, *J* = 9.3 Hz, 2H, Ar), 7.62 (d, *J* = 7.3, 4H, Ar), 8.00 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR δ 20.9 (-<u>C</u>H$_3$), 44.4 (<u>C</u>H-Ar$_3$), 112.9, 117.6, 120.0, 121.0, 128.0, 128.2, 128.4, 128.6, 128.7, 128.8, 130.5, 134.1, 135.6, 136.5, 140.1 (<u>Ar</u>); MS (El) *m/z* 490 (M^+); HRMS calcd for $C_{34}H_{26}N_4$: 490.2157; found: 490.2167.

Bis(3-(4-methoxyphenyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3e**) (ST310)

To a THF solution (4 mL) of *N*-(2-hydroxyl-2-(4-methoxyphenyl)-1-(2-pyridyl)ethyl)-4-methoxy-benzenecarbothioamide (**1e**) (197 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-(4-methoxyphenyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3e**) (0.102 g, 0.018 mmol, 72% per 0.25 mmol) as a green solid; mp 101-102 °C; IR (KBr) 3343, 3070, 3000, 2934, 2836, 2047, 1747, 1662, 1607, 1528, 1509, 1461, 1437, 1364, 1303, 1251, 1173, 1110, 1083, 1027, 964, 836, 791, 743, 699, 596, 515, 444, 416 cm^{-1}; ^1H NMR ($CDCl_3$) δ 3.65 (s, 3H, -OC<u>H</u>$_3$), 3.73 (s, 6H, -OC<u>H</u>$_3$), 6.21 (s, 1H, C<u>H</u>-Ar$_3$), 6.30 (ddd, *J* = 7.3, 6.3, 1.0 Hz, 2H, Ar), 6.39 (dd, *J* = 9.3, 6.3 Hz, 2H, Ar), 6.73 (d, *J* = 8.8 Hz, 2H, Ar), 6.89 (d, *J* = 8.8 Hz, 4H, Ar), 7.29 (d, *J* = 9.3 Hz, 2H, Ar), 7.31 (d, *J* = 8.8 Hz, 2H, Ar), 7.57 (d, *J* = 8.8 Hz, 4H, Ar), 7.95 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR δ 44.0 (<u>C</u>H-Ar$_3$), 55.1 (-O<u>C</u>H$_3$), 55.2 (-O<u>C</u>H$_3$), 112.7, 113.5, 114.2, 117.3, 119.6, 121.0, 122.9, 128.0, 129.6, 129.7, 133.7, 135.3, 136.5, 158.0, 159.7 (<u>Ar</u>); MS (El) *m/z* 560 (M^+); HRMS calcd for $C_{36}H_{30}N_4$: 566.2318, found: 560.2308.

Bis(3-(2-pyridyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3f**) (ST338)

To a THF solution (4 mL) of *N*-(2-hydroxy-2-(4-methoxyphenyl)-1-(2-pyridyl)ethyl)-2-pyridinecarbothioamide (**1f**) (366 mg, 1.0 mmol) was added I_2 (761 mg, 3.0 mmol) and pyridine (0.244 mL, 3.0 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture

was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1 – 1 : 1) with shielding of light to give bis(3-(2-pyridyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3f**) (0.137 g, 0.027 mmol, 54% per 0.50 mmol) as a green solid; mp 188-189 °C; IR (KBr) 3356, 3120, 2929, 2361, 2335, 1905, 1839, 1792, 1753, 1675, 1584, 1560, 1508, 1427, 1367, 1308, 1243, 1179, 1147, 1128, 1111, 1070, 1033, 1006, 964, 911, 886, 828, 798, 752, 696, 616, 570, 518, 469, 452, 442, 419 cm^{-1}; ^1H NMR δ 3.63 (s, 3H, -OC$\underline{H}_3$), 6.25 (s, 1H, C$\underline{H}$-Ar_3), 6.52 (ddd, *J* = 7.8, 6.3, 1.5 Hz, 2H, Ar), 6.58 (ddd, *J* = 9.8, 6.3, 1.0 Hz, 2H, Ar), 6.71 (d, *J* = 8.8 Hz, 2H, Ar), 6.97 (ddd, *J* = 7.3, 4.9, 1.0 Hz, 2H, Ar), 7.24 (d, *J* = 7.8 Hz, 2H, Ar), 7.45 (d, *J* = 8.8 Hz, 2H, Ar), 7.55 (td, *J* = 7.3, 2.0 Hz, 2H, Ar), 8.18 (d, *J* = 9.3Hz, 2H, Ar), 8.45 (ddd, *J* = 4.9, 2.0, 1.0 Hz, 2H, Ar), 9.77 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR: 55.1 (-O$\underline{C}H_3$), δ 44.0 ($\underline{C}$H-Ar_3), 113.4, 113.5, 118.8, 119.1, 121.1, 122.0, 125.7, 129.7, 130.0, 133.6, 134.6, 135.0, 136.2, 148.0, 151.2, 158.0 (Ar); MS (El) *m/z* 508 (M^+); HRMS calcd for $C_{32}H_{24}N_6O$: 508.2012, found: 508.2003.

Bis(3-(2-pyridyl)-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3g**) (ST339)

To a THF solution (4 mL) of *N*-(2-hydroxy-2-(4-chlorophenyl)-1-(2-pyridyl)ethyl)-2-pyridinecarbothioamide (**1g**) (185 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1 – 1 : 1) with shielding of light to give bis(3-(2-pyridyl)-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3g**) (0.0992 g, 0.019 mmol, 77% per 0.25 mmol) as a green solid; mp 81-82 °C; IR (KBr) 3303, 3115, 3015, 2926, 2855, 1903, 1754, 1714, 1665, 1631, 1588, 1561, 1504, 1427, 1337, 1310, 1375, 1193, 1147, 1127, 1089, 1045, 1013, 965, 925, 753, 710, 666, 616, 592, 540, 528, 503, 453, 424, 402 cm^{-1}; ^1H NMR δ 6.25 (s, 1H, C$\underline{H}$-Ar_3), 6.54 (ddd, *J* = 7.3, 6.8, 1.0 Hz, 2H, Ar), 6.61 (ddd, *J* = 9.3, 6.3, 1.0 Hz, 2H, Ar), 6.69 (ddd, *J* = 7.3, 4.9, 1.0 Hz, 2H, Ar), 7.14 (d, *J* = 8.6 Hz, 2H, Ar), 7.25 (d, *J* = 8.6 Hz, 2H, Ar), 7.47 (d, *J* = 9.3 Hz, 2H, Ar), 7.58 (ddd, *J* = 8.3, 7.3, 2.0 Hz, 2H, Ar), 8.18 (d, *J* = 8.3Hz, 2H, Ar), 8.46 (dd, *J* = 4.9, 1.0 Hz, 2H, Ar), 9.78 (d, *J* = 7.3 Hz, 2H, Ar); ^{13}C NMR δ 44.1 ($\underline{C}$H-Ar_3), 113.5, 118.6, 119.4, 121.2, 122.0, 125.8, 128.2, 130.0, 130.1, 132.0, 133.7, 133.8, 136.3, 141.4, 148.0, 151.2 (Ar); MS (El) *m/z* 512 (M^+); HRMS calcd for $C_{31}H_{21}ClN_6$: 512.1516,

found: 512.1519.

Bis(3-(2-thienyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3h**) (ST355)

To a THF solution (4 mL) of *N*-(2-(4-methoxyphenyl)-2-hydroxy-1-(2-pyridyl)ethyl)-2-thiophenecarbothioamide (**1h**) (185 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-(2-thienyl)-1-imidazo[1,5-a]pyridyl)-4-methoxyphenylmethane (**3h**) (0.074 g, 0.014 mmol, 57% per 0.25 mmol) as a green solid; mp 87-88 °C; IR (KBr) 3325, 3098, 2930, 2834, 1745, 1710, 1681, 1600, 1509, 1478, 1432, 1412, 1355, 1301, 1247, 1176, 1113, 1084, 1032, 998, 913, 845, 797, 732, 699, 618, 564, 531, 452, 412 cm^{-1}; ^1H NMR δ 3.65 (s, 3H, -OC<u>H</u>$_3$), 6.18 (s, 1H, C<u>H</u>-Ar$_3$), 6.44 (dd, *J* = 6.8, 6.3 Hz, 2H, Ar), 6.61 (dd, *J* = 8.3, 6.3 Hz, 2H, Ar), 6.72 (d, *J* = 8.8, 2H, Ar), 7.02 (dd, *J* = 4.9, 3.4 Hz, 2H, Ar), 7.24 (dd, *J* = 4.9, 1.0 Hz, 2H, Ar), 7.28 (d, *J* = 8.3 Hz, 2H, Ar), 7.35 (dd, *J* = 3.4, 1.0 Hz, 2H, Ar), 7.52 (d, *J* = 8.8 Hz, 2H, Ar), 8.10 (d, *J* = 6.8Hz, 2H, Ar); ^{13}C NMR δ 44.8, 44.9 (<u>C</u>H-Ar$_3$), 55.1, 55.2 (-O<u>C</u>H$_3$), 113.5, 117.7, 119.9, 120.0, 121.3, 121.4, 124.5, 124.6, 125.5, 125.6, 127.5, 128.7, 129.5, 129.6, 131.2, 132.8, 134.4, 135.1, 158.0 (Ar); MS (El) *m/z* 518 (M^+); HRMS calcd for $C_{30}H_{22}N_4OS_2$: 518.1235, found: 518.1213.

Bis(3-(thienyl)-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3i**) (ST356)

To a THF solution (4 mL) of *N*-(2-(4-chlorophenyl)-2-hydroxy-1-(2-pyridyl)ethyl)-2-thiophenecarbothioamide (**1i**) (186 mg, 0.5 mmol) was added I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give bis(3-(thienyl)-1-imidazo[1,5-a]pyridyl)-4-chlorophenylmethane (**3i**) (0.082 g, 0.016 mmol, 62% per 0.25 mmol) as a green solid; mp 90-91 °C; IR (KBr) 3434, 3070, 2930, 2366, 2324, 1899, 1734, 1630, 1588, 1511, 1487, 1404, 1332, 1304, 1260, 1236, 1170, 1112, 1088, 1014, 933, 913, 845, 781, 701, 614, 494, 454, 425 cm^{-1}; ^1H NMR δ 6.18 (s, 1H, C<u>H</u>-Ar$_3$), 6.43 (ddd, *J*

= 6.8, 6.3, 1.5 Hz, 2H, Ar), 6.51 (dd, *J* = 9.3, 6.3 Hz, 2H, Ar), 7.00 (dd, *J* = 4.9, 3.4 Hz, 2H, Ar, 2H, Ar), 7.12 (d, *J* = 8.3 Hz, 2H, Ar), 7.24 (d, *J* = 4.9 Hz, 2H, Ar), 7.28 (d, *J* = 8.3 Hz, 2H, Ar), 7.33 (d, *J* = 3.4 Hz, 2H, Ar), 7.55 (d, *J* = 9.3 Hz, 2H, Ar), 8.09 (d, *J* = 6.8 Hz, 2H, Ar); ^{13}C NMR δ 43.8 (C<u></u>H-Ar$_3$), 113.6, 118.0, 119.6, 121.4, 124.6, 125.6, 127.5, 128.2, 126.7, 130.0, 131.3, 131.9, 132.6, 133.5, 141.5 (Ar); MS (El) *m/z* 522 (M^+); HRMS calcd for $C_{29}H_{19}ClN_4S_2$: 522.0740, found: 522.0737.

[3-(4-Methoxyphenyl)-1-imidazo[1,5-a]pyridyl](3-phenyl-1-imidazo[1,5-a]pyridyl)phenylmethane (**6a**) (ST349)

To a THF solution (4 mL) of *N*-(2-hydroxy-2-phenyl-1-(2-pyridyl)ethyl)benzenecarbothioamide (**2f**) (167 mg, 0.5 mmol) was added 3-(4-methoxyphenyl)imidazo[1,5-a]pyridine (**5a**) (112 mg, 0.5 mmol), I_2 (381 mg, 1.5 mmol) and pyridine (0.121 mL, 1.5 mmol) at room temperature. The mixture was stirred at room temperature for 30 min. The reaction mixture was poured onto $Na_2S_2O_3$ (aq) and extracted with CH_2Cl_2. The organic layer was dried over $MgSO_4$ and concentrated *in vacuo*. The residue was purified by column chromatography on silica gel (*n*-hexane : ethyl acetate = 2 : 1) with shielding of light to give [3-(4-methoxyphenyl)-1-imidazo[1,5-a]pyridyl](3-phenyl-1-imidazo[1,5-a]pyridyl)phenylmethane (**6a**) (0.161 g, 0.032 mmol, 64% per 0.50 mmol) as a green solid; mp 74-75 °C; IR (KBr) 3063, 3024, 2936, 2836, 2045, 1898, 1748, 1655, 1631, 1610, 1573, 1529, 1493, 1461, 1405, 1365, 1303, 1289, 1250, 1216, 1172, 1110, 1074, 1030, 1001, 963, 916, 836, 795, 749, 698, 666, 617, 597, 578, 514, 423 cm^{-1}; ^{1}H NMR ($CDCl_3$) δ 3.69 (s, 3H, -OC<u></u>H$_3$), 6.26 (s, 1H, C<u></u>H-Ar$_3$), 6.24-6.30 (m, 2H, Ar), 6.34-6.42 (m, 2H, Ar), 6.87 (d, *J* = 8.8 Hz, 2H, Ar), 7.08 (d, *J* = 7.3 Hz, 1H, Ar), 7.16 (t, *J* = 7.3 Hz, 2H, Ar), 7.24 (d, *J* = 7.3 Hz, 1H, Ar), 7.33(t, *J* = 7.3 Hz, 2H, Ar), 7.34 (d, *J* = 6.3 Hz, 1H, Ar), 7.36 (d, *J* = 8.8 Hz, 1H, Ar), 7.39 (d, *J* = 7.3 Hz, 2H, Ar), 7.55 (d, *J* = 8.8 Hz, 2H, Ar), 7.64 (d, *J* = 7.3 Hz, 2H, Ar), 7.92 (d, *J* = 7.3 Hz, 1H, Ar), 8.01 (d, *J* = 7.3 Hz, 1H, Ar); ^{13}C NMR δ 44.9 (C<u></u>H-Ar$_3$), 55.2 (-OC<u></u>H$_3$), 112.6, 112.8, 114.2, 117.3, 117.6, 119.61, 119.63, 120.96, 120.99, 123.0, 126.1, 128.0, 128.1, 128.2, 128.5, 128.7, 128.8, 129.5, 130.5, 133.4, 134.0, 136.6, 143.2, 159.6 (<u>Ar</u>); MS (El) *m/z* 506 (M^+); HRMS calcd for $C_{34}H_{26}N_4O$: 506.2107, found: 506.2107.

X-ray Structure Analysis. The measurement was carried out on a Rigaku/MSC Mercury CCD diffractometer with graphite-monochlomated Mo Kα radiation (λ = 0.71069 Å). The structures were solved and refined using the teXsan® crystallographic softwere package of Molecular Structure Corporation. The X-ray quality crystal was obtained by slow diffusion of hexane and CH_2Cl_2 into **3g** (0.23 mg). The crystal was cut from the grown crystal and mounted on a glass fiber. The structure was solved by direct method using SHELXL-97. Scattering factors for neutral atoms were from Cromer and Waber, and anomalous dispersion effect were used. The function minimized was $\Sigma w(F_o^2-F_c^2)^2$ and the weighting scheme employed $w = [\sigma^2_c(F_o^2)+(p(\max(F_o^2, 0)+2F_c^2/3)^2)]^{-1}$. A full-matrix least-squares refinement was executed with non-hydrogen atoms being anisotropic. The final least square cycle included fixed hydrogen atoms at calculated positions of which each isotropic thermal parameter was set to 1.2 times of that of the connecting atom. Crystal data and measurement description are summarized in Table 3.

Table 3. Crystal data and structure refinement for **3g**.

Identification code	crystalclear	
Empirical formula	$C_{31}H_{21}ClN_6$	
Formula weight	512.99	
Temperature	193(2) K	
Wavelength	0.71070 Å	
Crystal system	Monoclinic	
Space group	$P\,2_1/c$	
Unit cell dimensions	a = 11.262(6) Å	α = 90°.
	b = 10.562(6) Å	β = 96.078(7)°.
	c = 20.766(12) Å	γ = 90°.
Volume	2456(2) $Å^3$	
Z	4	
Density (calculated)	1.387 mg/m^3	
Absorption coefficient	0.190 mm^{-1}	
F(000)	1064	
Crystal size	0.30 x 0.20 x 0.20 mm^3	
Theta range for data collection	3.19 to 27.50°.	
Index ranges	-14<=h<=14, -13<=k<=13, -17<=l<=26	

Reflections collected	19735
Independent reflections	5616 [R(int) = 0.0806]
Completeness to theta = 27.50°	99.5 %
Absorption correction	Integration
Max. and min. transmission	0.989 and 0.950
Refinement method	Full-matrix least-squares on F^2
Data / restraints / parameters	5616 / 0 / 347
Goodness-of-fit on F^2	1.000
Final R indices [I>2sigma(I)]	R1 = 0.1074, wR2 = 0.2353
R indices (all data)	R1 = 0.1447, wR2 = 0.2600
Largest diff. peak and hole	0.267 and –0.310 e.$Å^{-3}$