Supporting Information

A Facile Synthesis of Tetrahydroimidazo[1,2-a]pyridines and Tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridines through NHC-Catalyzed Cascade Annulations

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1 General considerations

Common reagents and materials were purchased from commercial sources and purified by distillation or recrystalization. Melting points were determined in open capillaries and were uncorrected. IR spectra were taken on a FT-IR-Tensor 27 spectrometer in KBr pellets and reported in cm⁻¹. ¹H NMR spectra were measured on a Bruker DPX 400 MHz spectrometer in DMSO-d₆ (100 MHz, ¹³C NMR) or CDCl₃ with chemical shift (δ) given in ppm relative to TMS as internal standard. High-resolution mass spectra (HRMS) were obtained on a micrOTOF-Q II HRMS/MS instrument (Bruker) with the technique of electrospray ionization.

2. Experimental
General Procedure for the Synthesis of \(3\): Into an oven-dried 25 mL vial were weighed precatalyst \(4d\) (34 mg) and \(\text{Cs}_2\text{CO}_3\) (520 mg). Toluene (7 mL) was added to the mixture. The resulting mixture was stirred at 25 °C for 5 min followed by the addition of \(\alpha\)-bromo-\(\alpha,\beta\)-unsaturated aldehyde (1.5 mmol) and HKA 2 (1 mmol) after the reaction mixture was stirred for about 7 min, the reaction system was kept at 65 °C with stirring until completion (monitored by TLC). After removal of the solvent under reduced pressure, the crude product was purified by column chromatography (silica gel, mixtures of ethyl acetate / petroleum ether, 3:1, v/v).

General Procedure for the Synthesis of \(5\): Into an oven-dried 25 mL vial were weighed catalyst \(4d\) (34 mg) and \(\text{Cs}_2\text{CO}_3\) (520 mg). Toluene (7 mL) was added to the mixture. The resulting mixture was stirred at 25 °C for 5 min followed by the addition of \(\alpha\)-bromo-\(\alpha,\beta\)-unsaturated aldehyde (1.5 mmol) and HKA 2 (1 mmol) after the reaction mixture was stirred for about 7 min, the reaction system was kept at 65 °C with stirring until completion (monitored by TLC). After the removal of the solvent under reduced pressure, \(\text{K}_2\text{CO}_3\) (138 mg, 1.0 mmol) and DMF (9 mL) was added and the mixture was heated to 100 °C. After the completion of the reaction as indicated by TLC, the mixture was cooled to room temperature. An amount of 50 mL water was added to precipitate the product, which was then filtered and washed with small amount of ethanol to give the pure product \(5\).

3. Spectral data for all compounds

**Compound 3a:** 8-benzoyl-7-phenyl-2,3,6,7-tetrahydroimidazo[1,2-\(a\)]pyridin-5(1H)-one
White solid; mp: 165-167 °C (reported 166-167 °C); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 9.74 (s, 1H), 7.31 (d, \(J = 7.2\) Hz, 1H), 7.27 – 7.15 (m, 7H), 7.02 (d, \(J = 8.0\) Hz, 2H), 4.14 – 4.07 (m, 2H), 3.96 – 3.83 (m, 3H), 2.97 (dd, \(J_1 = 6.8\) Hz, \(J_2 = 16.4\) Hz, 1H), 2.78 (dd, \(J_1 = 2.0\) Hz, \(J_2 = 16.4\) Hz, 1H); IR (potassium bromide) (\(\nu\), cm\(^{-1}\)): 3267, 2977, 2902, 1694, 1633, 1518, 1485, 1370, 1314, 1207, 1014, 721, 700; HRMS (ESI): m/z Calcd. for C\(_{20}\)H\(_{17}\)N\(_2\)O\(_2\) [M-H]: 317.1290, found: 317.1290.

**Compound 3b:**
7-(4-chlorophenyl)-8-(2,4-dichlorobenzoyl)-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

White solid; mp: 168-170 °C; \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 9.56 (s, 1H), 7.37 (s, 1H), 7.17 (d, \(J = 7.6\) Hz, 2H), 7.07 (s, 1H), 6.82 (d, \(J = 7.6\) Hz, 2H), 6.68 (s, 1H), 4.20 – 4.11 (m, 1H), 4.03 – 3.89 (m, 3H), 3.76 (d, \(J = 6.0\) Hz, 1H), 3.04 (dd, \(J_1 = 7.2\) Hz, \(J_2 = 16.8\) Hz, 1H), 2.73 (d, \(J = 16.8\) Hz, 1H); IR (potassium bromide) (\(\nu\), cm\(^{-1}\)): 3230, 2987, 1685, 1635, 1535, 1486, 1372, 1319, 822, 789; HRMS (ESI): m/z Calcd. for C\(_{20}\)H\(_{14}\)Cl\(_3\)N\(_2\)O\(_2\) [M-H]: 419.0121, found: 419.0108.

**Compound 3c:**
9-benzoyl-8-phenyl-3,4,7,8-tetrahydro-1H-pyrido[1,2-a]pyrimidin-6(2H)-one

Flaxen solid; mp: 168-170 °C (reported 167-168 °C); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 12.99 (s, 1H), 7.26 – 7.19 (m, 6H), 7.10 (d, \(J = 7.2\) Hz, 2H), 7.05 (d, \(J = 6.8\) Hz, 2H), 4.04 – 3.95 (m, 2H), 3.58 – 3.48 (m, 3H), 2.98 – 2.93 (m, 1H), 2.82 (d, \(J = 15.6\) Hz, 1H), 2.07 (s, 2H); IR (potassium bromide) (\(\nu\), cm\(^{-1}\)): 3562, 3023, 1696, 1617, 1146, 722, 700; HRMS (ESI): m/z Calcd. for C\(_{21}\)H\(_{19}\)N\(_2\)O\(_2\) [M-H]: 331.1447, found: 331.1448.

**Compound 3d:**
8-benzoyl-7-(4-chlorophenyl)-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Ash colored solid; mp: 170-172 °C (reported 190-191 °C); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\) 9.73 (s, 1H), 7.36 – 7.23 (m, 5H), 7.16 (d, \(J = 7.2\) Hz, 2H), 6.98 (d, \(J = 8.0\) Hz, 2H), 4.17 – 4.10 (m, 2H, CH\(_2\)), 3.98 – 3.89 (m, 3H), 2.99 (dd, \(J_1 = 6.8\) Hz, \(J_2 = 16.4\) Hz, 1H), 2.75 (d, \(J = 16.0\) Hz, 1H); \(^{13}\)C NMR (100 MHz, CDCl\(_3\)) \(\delta\) 192.1, 167.7, 157.3, 142.7, 141.2, 132.5, 129.3, 128.9, 128.1, 128.0, 126.3, 88.0, 42.8, 41.9, 40.7, 37.7; IR (potassium bromide) (\(\nu\), cm\(^{-1}\)): 3297, 3051, 2987, 1685, 1635, 1486, 1372, 1319, 822, 789; HRMS (ESI): m/z Calcd. for C\(_{20}\)H\(_{16}\)ClN\(_2\)O\(_2\) [M-H]: 351.0900, found: 351.0901.
**Compound 3e:**
8-(2,4-dichlorobenzoyl)-7-phenyl-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

![Compound 3e](image)

White solid; mp: 209-210 °C; $^1$H NMR (400 MHz, CDCl$_3$) δ 9.55 (s, 1H), 7.42 – 7.35 (m, 1H), 7.16 (s, 3H), 7.01 (s, 1H), 6.86 (s, 2H), 6.65 (s, 1H), 4.21 – 4.07 (m, 1H), 4.00 – 3.89 (m, 3H), 3.75 (d, $J$ = 4.0 Hz, 1H, CH), 3.03 (dd, $J_1$ = 6.8 Hz, $J_2$ = 16.4 Hz, 1H), 2.75 (d, $J$ = 16.4 Hz, 1H); IR (potassium bromide) (v, cm$^{-1}$): 3278, 3072, 1687, 1639, 1533, 1479, 1446, 1371, 1197, 1156, 1010, 843, 701, 182; HRMS (ESI) m/z Calcd. for C$_{20}$H$_{15}$Cl$_2$N$_2$O$_2$ [M-H]$: 385.0511, found: 385.0494.

**Compound 3f:**
7-(3-chlorophenyl)-8-(2,4-dichlorobenzoyl)-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

![Compound 3f](image)

White solid; mp: 211-213 °C; $^1$H NMR (400 MHz, CDCl$_3$) δ 9.57 (s, 1H), 7.35 (s, 1H), 7.15 – 7.06 (m, 3H), 6.81 (s, 1H), 6.75 (d, $J$ = 6.8 Hz, 1H), 6.68 (s, 1H), 4.18 – 4.09 (m, 1H), 4.02 – 3.90 (m, 3H), 3.75 (d, $J$ = 6.4 Hz, 1H, CH), 3.03 (dd, $J_1$ = 7.6 Hz, $J_2$ = 16.8 Hz, 1H), 2.74 (dd, $J_1$ = 2.0 Hz, $J_2$ = 16.8 Hz, 1H); IR (potassium bromide) (v, cm$^{-1}$): 3318, 3045, 1691, 1631, 1586, 1518, 1487, 1568, 1368, 1204, 1021, 822, 777; HRMS (ESI) m/z: Calcd. for C$_{20}$H$_{14}$Cl$_3$N$_2$O$_2$ [M-H]$: 419.0121, found: 419.0121.

**Compound 3g:**
7-(4-chlorophenyl)-8-(2,5-dichlorobenzoyl)-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

![Compound 3g](image)

Ash colored solid; mp: 216-218 °C; $^1$H NMR (400 MHz, CDCl$_3$) δ 9.52 (s, 1H), 7.24 (s, 1H), 7.19 (d, $J$ = 2.4 Hz, 1H), 7.16 (d, $J$ = 8.0 Hz, 2H), 6.76 (d, $J$ = 8.4 Hz, 2H), 6.64 (s, 1H), 4.18 – 4.07 (m, 1H), 4.01 – 3.87 (m, 3H), 3.74 (d, $J$ = 4.0 Hz, 1H, CH), 3.03 (dd, $J_1$ = 7.6 Hz, $J_2$ = 16.8 Hz, 1H), 2.70 (dd, $J_1$ = 1.6 Hz, $J_2$ = 16.4 Hz, 1H); $^{13}$C NMR (100 MHz, CDCl$_3$) δ 167.4, 157.2, 141.2,
Compound 3h:
8-(2-chlorobenzoyl)-7-phenyl-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Yellow solid; mp: 175-176 °C; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 9.57 (s, 1H), 7.33 (s, 1H), 7.24 – 7.16 (m, 4H), 7.04 (s, 1H), 6.84 (d, $J = 6.0$ Hz, 2H), 6.72 (s, 1H), 4.19 – 4.08 (m, 1H), 3.95 – 3.86 (m, 3H), 3.80 (d, $J = 4.4$ Hz, 1H, CH), 3.06 (dd, $J_1 = 7.2$ Hz, $J_2 = 16.4$ Hz, 1H), 2.76 (d, $J = 16.8$ Hz, 1H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 167.9, 156.9, 143.9, 140.1, 129.5, 128.5, 127.8, 126.7, 126.5, 89.2, 42.8, 41.9, 40.5, 38.0; IR (potassium bromide) ($v$, cm$^{-1}$): 3298, 3058, 3027, 1687, 1636, 1531, 1485, 1449, 1372, 1208, 1163, 1072, 1017, 936, 762, 733, 691, 633; HRMS (ESI): m/z Calcd. for C$_{20}$H$_{16}$ClN$_2$O$_2$ [M-H]: 351.0900, found: 351.0891.

Compound 3i:
8-(4-bromobenzoyl)-7-phenyl-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Yellow solid; mp: 189-190 °C; $^1$H NMR (400 MHz, CDCl$_3$) $\delta$ 9.75 (s, 1H), 7.38 (d, $J = 7.6$ Hz, 2H), 7.31 – 7.21 (m, 3H), 7.05 (d, $J = 7.6$ Hz, 4H), 4.18 – 4.10 (m, 1H), 4.07 (d, $J = 6.8$ Hz, 1H), 3.99 – 3.87 (m, 3H), 2.99 (dd, $J_1 = 6.8$ Hz, $J_2 = 16.4$ Hz, 1H), 2.80 (d, $J = 16.4$ Hz, 1H); $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta$ 190.6, 167.9, 157.7, 143.9, 140.1, 131.1, 128.9, 128.3, 126.9, 126.7, 123.4, 88.2, 42.9, 41.9, 40.9, 38.2; IR (potassium bromide) ($v$, cm$^{-1}$): 3282, 3061, 1687, 1637, 1530, 1443, 1366, 1316, 1202, 1153, 1006, 767; HRMS (ESI): m/z Calcd. for C$_{20}$H$_{16}$BrN$_2$O$_2$ [M-H]: 395.0395, found: 395.0406.
**Compound 3j:**
8-(2,4-dichlorobenzoyl)-7-(4-methoxyphenyl)-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Yellow solid; mp: 127-129 °C; ¹H NMR (400 MHz, CDCl₃) δ 9.52 (s, 1H), 7.34 (s, 1H), 7.03 (d, J = 5.2 Hz, 1H), 6.77 (d, J = 8.8 Hz, 2H), 6.71 (d, J = 8.8 Hz, 2H), 6.66 (s, 1H), 4.17 – 4.10 (m, 1H), 3.95 – 3.88 (m, 3H), 3.75 (s, 3H), 3.70 (d, J = 6.8 Hz, 1H, CH), 2.99 (dd, J₁ = 2.0 Hz, J₂ = 16.4 Hz, 1H), 2.72 (dd, J₁ = 2.0 Hz, J₂ = 16.4 Hz, 1H); ¹³C NMR (100 MHz, CDCl₃) δ 188.6, 167.9, 158.4, 157.2, 138.6, 135.7, 134.6, 129.2, 128.7, 127.5, 126.8, 113.9, 89.3, 55.2, 42.8, 41.9, 40.8, 37.2; IR (potassium bromide) (v, cm⁻¹): 3327, 3076, 2933, 1738, 1690, 1638, 1529, 1486, 1373, 1322, 1232, 1021, 826, 658, 564; HRMS (ESI): m/z Calcd. for C₂₁H₁₇Cl₂N₂O₃ [M-H]: 415.0616, found: 415.0600.

**Compound 3k:**
8-benzoyl-7-methyl-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Brown solid; mp: 148-150 °C; ¹H NMR (400 MHz, CDCl₃) δ 9.51 (s, 1H), 7.40 (s, 5H), 4.11 – 4.04 (m, 1H), 3.93 – 3.75 (m, 3H), 3.06 – 3.00 (m, 1H, CH), 2.72 (dd, J₁ = 6.4 Hz, J₂ = 16.4 Hz, 1H), 2.41 (dd, J₁ = 1.6 Hz, J₂ = 16.4 Hz, 1H), 0.95 (d, J = 6.8 Hz, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 186.6, 163.7, 150.9, 136.4, 123.7, 122.9, 121.1, 86.2, 37.4, 36.6, 34.5, 22.2, 16.9; IR (potassium bromide) (v, cm⁻¹): 3288, 2958, 1693, 1624, 1524, 1491, 1377, 1205, 1019, 748, 700, 643; HRMS (ESI): m/z Calcd. for C₁₅H₁₅N₂O₂ [M-H]: 255.1134, found: 255.1142.

**Compound 3l:**
8-benzoyl-7,7-dimethyl-2,3,6,7-tetrahydroimidazo[1,2-a]pyridin-5(1H)-one

Yellow solid; mp: 125-127 °C; ¹H NMR (400 MHz, CDCl₃) δ 8.05 (d, J = 7.6 Hz, 2H), 7.63 (t, J = 7.2 Hz, 1H), 7.51 (t, J = 7.6 Hz, 2H), 4.59 (s, 1H), 3.99 – 3.79 (m, 4H), 3.05 (d, J = 17.2 Hz, 1H), 2.28 (t, J = 16.8 Hz, 1H), 1.16 (s, 3H), 1.02 (s, 3H); ¹³C NMR (100 MHz, CDCl₃) δ 191.9, 162.4,
Compound 5a:
9-chloro-6-(4-methoxyphenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione

White solid; mp: 220-222 °C; $^1$H NMR (400 MHz, DMSO-d$_6$) $\delta$ 8.02 (s, 1H), 7.74 (d, $J$ = 8.4 Hz, 1H), 7.55 (d, $J$ = 8.8 Hz, 1H), 7.15 (d, $J$ = 8.0 Hz, 2H), 6.80 (d, $J$ = 8.0 Hz, 2H), 4.55 – 4.37 (m, 3H), 4.23 – 4.10 (m, 2H), 3.68 (s, 3H), 3.12 (dd, $J_1$ = 8.0 Hz, $J_2$ = 16.8 Hz, 1H), 2.69 (d, $J$ = 16.4 Hz, 1H); IR (potassium bromide) (v, cm$^{-1}$): 3061, 2907, 1709, 1635, 1546, 1514, 1381, 1251, 1175, 1035, 829, 793, 507; HRMS (ESI): m/z Calcd. for C$_{21}$H$_{17}$ClN$_2$O$_3$ [M+Na]$^+$: 403.0825, found: 403.0823.

Compound 5b:
10-chloro-6-(4-methoxyphenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione

White solid; mp: 154-155 °C (reported 224-226 °C$^2$); $^1$H NMR (400 MHz, DMSO-d$_6$) $\delta$ 8.07 (d, $J$ = 8.4 Hz, 1H), 7.64 (s, 1H), 7.35 (d, $J$ = 8.8 Hz, 1H), 7.15 (d, $J$ = 8.0 Hz, 2H), 6.80 (d, $J$ = 8.0 Hz, 2H), 4.54 – 4.36 (m, 3H), 4.22 – 4.09 (m, 2H), 3.12 (dd, $J_1$ = 8.0 Hz, $J_2$ = 16.8 Hz, 1H), 2.68 (d, $J$ = 16.4 Hz, 1H); IR (potassium bromide) (v, cm$^{-1}$): 3059, 3036, 1706, 1638, 1613, 1573, 1540, 1513, 1434, 1372, 1247, 1029, 848, 783; HRMS (ESI): m/z Calcd. for C$_{21}$H$_{17}$ClN$_2$NaO$_3$ [M+Na]$^+$: 403.0825, found: 403.0829.
**Compound 5c:**
9-chloro-6-(3-methoxyphenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione

Straw-colored solid; mp: 288–290 °C; \(^1\)H NMR (400 MHz, DMSO-\(d_6\)) \(\delta\) 8.02 (d, \(J = 2.0\) Hz, 1H), 7.73 (dd, \(J_1 = 2.4\) Hz, \(J_2 = 8.8\) Hz, 1H), 7.53 (d, \(J = 8.8\) Hz, 1H), 7.16 (t, \(J = 8.0\) Hz, 1H), 6.82 – 6.75 (m, 3H), 4.55 – 4.47 (m, 2H), 4.42 – 4.35 (m, 1H, CH), 4.23 – 4.11 (m, 2H), 3.69 (s, 3H), 3.16 (dd, \(J_1 = 8.4\) Hz, \(J_2 = 16.8\) Hz, 1H), 2.73 (d, \(J = 16.8\) Hz, 1H); \(^13\)C NMR (100 MHz, DMSO-\(d_6\)) \(\delta\) 172.2, 167.6, 159.3, 147.9, 145.0, 135.5, 131.4, 129.5, 127.6, 126.0, 124.7, 118.5, 117.9, 113.0, 111.3, 99.2, 54.9, 45.4, 42.4, 38.8, 34.0; IR (potassium bromide) (\(v,\) cm\(^{-1}\)): 3054, 2909, 2838, 1703, 1640, 1599, 1580, 1519, 1470, 1378, 1278, 1157, 1035, 808; HRMS (ESI): m/z Calcd. for C\(_{21}\)H\(_{17}\)ClN\(_2\)NaO\(_3\) [M\(+\)Na]\(^+\): 403.0825, found: 403.0820.

**Compound 5d:**
9-chloro-6-(3-chlorophenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione

Brown solid; mp: 262-264 °C; \(^1\)H NMR (400 MHz, DMSO-\(d_6\)) \(\delta\) 8.03 (d, \(J = 4.0\) Hz, 1H), 7.75 (dd, \(J_1 = 4.0\) Hz, \(J_2 = 8.0\) Hz, 1H), 7.56 (d, \(J = 8.0\) Hz, 1H), 7.33 – 7.20 (m, 4H), 4.52 (dd, \(J_1 = 8.0\) Hz, \(J_2 = 16.0\) Hz, 2H), 4.41 (dd, \(J_1 = 8.8\) Hz, \(J_2 = 17.2\) Hz, 1H), 4.23 – 4.13 (m, 2H), 3.18 (dd, \(J_1 = 8.0\) Hz, \(J_2 = 16.0\) Hz, 1H), 2.74 (d, \(J = 16.0\) Hz, 1H); IR (potassium bromide) (\(v,\) cm\(^{-1}\)): 3056, 2962, 2910, 1702, 1644, 1617, 1586, 1546, 1515, 1379, 1318, 1224, 1180, 808; HRMS (ESI): m/z Calcd. for C\(_{20}\)H\(_{16}\)Cl\(_2\)NaO\(_2\) [M+Na]\(^+\): 407.0330, found: 407.0345.

**Compound 5e:**
10-chloro-6-(3-chlorophenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione
Brown solid; mp: 156-158 °C; ¹H NMR (400 MHz, DMSO-d₆) δ 8.07 (d, J = 8.4 Hz, 1H), 7.65 (s, 1H), 7.37 – 7.19 (m, 5H), 4.50 (d, J = 7.2 Hz, 2H), 4.40 (dd, J₁ = 9.2 Hz, J₂ = 17.6 Hz, 1H), 4.23 – 4.12 (m, 2H), 3.18 (dd, J₁ = 8.4 Hz, J₂ = 16.4 Hz, 1H), 2.73 (d, J = 16.8 Hz, 1H); ¹³C NMR (100 MHz, DMSO-d₆) δ 172.3, 159.4, 151.5, 150.1, 140.9, 138.3, 138.1, 132.4, 129.7, 129.1, 128.3, 127.3, 123.7, 123.2, 116.0, 115.4, 99.0, 45.9, 44.2; IR (potassium bromide) (v, cm⁻¹): 3060, 2967, 2909, 1691, 1648, 1616, 1583, 1538, 1519, 1373, 1228, 1092, 778, 684; HRMS (ESI): m/z Calcd. for C₂₀H₁₄Cl₂N₂NaO₂ [M+Na]⁺: 407.0330, found: 407.0327.

**Compound 5f:**
10-chloro-6-(4-chlorophenyl)-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione

Brown solid; mp: 151-153 °C (reported 223-225 °C); ¹H NMR (400 MHz, DMSO-d₆) δ 8.07 (d, J = 8.8 Hz, 1H), 7.65 (s, 1H), 7.36 (d, J = 8.8 Hz, 1H), 7.30 (d, J = 8.0 Hz, 2H), 7.27 (d, J = 8.0 Hz, 2H), 4.54 – 4.48 (m, 2H), 4.40 (dd, J₁ = 9.2 Hz, J₂ = 17.6 Hz, 1H), 4.23 – 4.10 (m, 2H), 3.17 (dd, J₁ = 8.0 Hz, J₂ = 16.8 Hz, 1H), 2.70 (d, J = 17.2 Hz, 1H); IR (potassium bromide) (v, cm⁻¹): 3057, 3036, 1712, 1640, 1614, 1577, 1520, 1420, 1369, 1315, 1231, 1093, 1013, 876, 842, 783, 648, 501; HRMS (ESI): m/z Calcd. for C₂₀H₁₄Cl₂N₂NaO₂ [M+Na]⁺: 407.0330, found: 407.0342.

**Compound 5g:**
10-chloro-6-phenyl-1,2,5,6-tetrahydrobenzo[b]imidazo[1,2,3-ij][1,8]naphthyridine-4,7-dione
Brown solid; mp: 288-290 °C (reported 252-254 °C); \(^1\)H NMR (400 MHz, DMSO-\(d_6\)) \(\delta\) 8.07 (d, \(J = 8.8\) Hz, 1H), 7.65 (s, 1H), 7.35 (d, \(J = 8.4\) Hz, 1H), 7.25 (s, 4H), 7.18 (d, \(J = 4.4\) Hz, 1H), 4.55 – 4.48 (m, 2H), 4.40 (dd, \(J_1 = 8.8\) Hz, \(J_2 = 17.6\) Hz, 1H), 4.23 – 4.11 (m, 2H), 3.18 (dd, \(J_1 = 8.0\) Hz, \(J_2 = 16.4\) Hz, 1H), 2.71 (d, \(J = 16.8\) Hz, 1H); IR (potassium bromide) (\(v\), cm\(^{-1}\)): 3076, 2972, 2890, 1706, 1644, 1614, 1576, 1520, 1366, 1090, 950, 831, 740, 524; HRMS (ESI): m/z Calcd. for C\(_{20}\)H\(_{15}\)ClN\(_2\)O\(_2\) [M+Na\(^+\)]: 373.0720, found: 373.0725.


\(^1\)H NMR Spectrum (400 MHz, CDCl\(_3\)) of Compound 3a
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3b

$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3b
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3c
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3d

$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3d
**1H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3e**

![1H NMR Spectrum](image)

**13C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3e**

![13C NMR Spectrum](image)
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3f

$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3f
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3g

$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3g
$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3h
$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3h

$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3i
$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3i

$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3j
$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3j

$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3k
$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3k

$^1$H NMR Spectrum (400 MHz, CDCl$_3$) of Compound 3l
$^{13}$C NMR Spectrum (100 MHz, CDCl$_3$) of Compound 3I
$^1$H NMR Spectrum (400 MHz, DMSO-$d_6$) of Compound 5a

$^{13}$C NMR Spectrum (100 MHz, DMSO-$d_6$) of Compound 5a
$^1$H NMR Spectrum (400 MHz, DMSO-$d_6$) of Compound 5b

$^1$H NMR Spectrum (400 MHz, DMSO-$d_6$) of Compound 5c
$^{13}$C NMR Spectrum (100 MHz, DMSO-$d_6$) of Compound 5c

$^1$H NMR Spectrum (400 MHz, DMSO-$d_6$) of Compound 5d
$^{13}$C NMR Spectrum (100 MHz, DMSO-$d_6$) of Compound 5d
$^1$H NMR Spectrum (400 MHz, DMSO-$d_6$) of Compound 5e

$^{13}$C NMR Spectrum (100 MHz, DMSO-$d_6$) of Compound 5e
**1H NMR Spectrum (400 MHz, DMSO-d$_6$) of Compound 5f**

**1H NMR Spectrum (400 MHz, DMSO-d$_6$) of Compound 5g**