Supporting Information

I$_2$/H$_2$O$_2$ mediated synthesis and photophysical properties of imidazole-fused heterocycles via [4+1] cyclization approach

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1. General Information

Reagents, solvents, and analytical methods:
Unless otherwise noted, all reactions were carried out under an atmospheric atmosphere. All reagents were from commercial sources and used as received without further purification. All solvents were dried by standard techniques and distilled before use. Column chromatography was performed on silica gel (200-300 meshes) using petroleum ether (bp. 60~90 °C) and ethyl acetate as eluent. ¹H NMR spectra were recorded on a Bruker Avance operating for ¹H NMR at 400 MHz, ¹³C NMR at 101 MHz, and ¹⁹F NMR at 376 MHz. NMR spectral data were reported in ppm relative to tetramethylsilane (TMS) as internal standard and CDCl₃ (¹H NMR δ 7.27, ¹³C NMR δ 77.0), DMSO (¹H NMR δ 5.20, ¹³C NMR δ 39.52) as solvent. All coupling constants (J) are reported in Hz. The following abbreviations were used to describe peak splitting patterns when appropriate: s = singlet, d = doublet, dd = double doublet, ddd = double doublet of doublets, t = triplet, dt = double triplet, q = quartet, m = multiplet, br = broad. High-resolution mass spectra (HRMS) are produced on Q-Exactive Orbitrap HR-MS (Thermo Fisher Scientific, Waltham, Massachusetts). All the amines and aldehydes used here were commercially available. The raw material used in the reaction were purchased from Aladdin, Macklin, and so on.
2. Optimization of Reaction Conditions and Control experiment

Table S1. Optimization for generating 3b[a]

| Entry | I₂ (x mmol) | H₂O₂ (mmol) | Solvent (5 mL) | Time (h) | Temp. (°C) | Yield (%) | Yield (%) |[b] |
|-------|-------------|-------------|----------------|----------|------------|-----------|-----------|
| 1     | 0           | 3           | CH₃CN          | 1.2      | 70         | 0         | 0         |
| 2     | 0.2         | 3           | CH₃CN          | 1.2      | 70         | 25        | 48        |
| 3     | 0.3         | 3           | CH₃CN          | 1.2      | 70         | 19        | 58        |
| 4     | 0.4         | 3           | CH₃CN          | 1.2      | 70         | 12        | 69        |
| 5     | 0.5         | 3           | CH₃CN          | 1.2      | 70         | 0         | 76        |
| 6     | 0.6         | 3           | CH₃CN          | 1.2      | 70         | 0         | 72        |
| 7     | 0.5         | 2           | CH₃CN          | 1.2      | 70         | 7         | 69        |
| 8     | 0.5         | 4           | CH₃CN          | 1.2      | 70         | 3         | 67        |
| 9     | 0.5         | 5           | CH₃CN          | 1.2      | 70         | 5         | 64        |
| 10    | 0.5         | 6           | CH₃CN          | 1.2      | 70         | 5         | 62        |
| 11    | 0.5         | 3           | DMF            | 1.2      | 70         | 0         | 75        |
| 12    | 0.5         | 3           | DMSO           | 1.2      | 70         | 0         | 74        |
| 13    | 0.5         | 3           | THF            | 1.2      | 70         | 32        | 47        |
| 14    | 0.5         | 3           | Dioxane        | 1.2      | 70         | 29        | 32        |
| 15    | 0.5         | 3           | DCE            | 1.2      | 70         | 48        | 39        |
| 16    | 0.5         | 3           | H₂O            | 1.2      | 70         | 0         | 35        |
| 17    | 0.5         | 3           | CH₃CH₂OH       | 1.2      | 70         | 10        | 73        |
| 18    | 0.5         | 3           | CH₃CN          | 0.5      | 70         | 11        | 69        |
| 19    | 0.5         | 3           | CH₃CN          | 1        | 70         | 7         | 77        |
| 20    | 0.5         | 3           | CH₃CN          | 2        | 70         | 0         | 84        |
| 21    | 0.5         | 3           | CH₃CN          | 3        | 70         | 0         | 61        |
| 22    | 0.5         | 3           | CH₃CN          | 4        | 70         | 0         | 54        |
| 23    | 0.5         | 3           | CH₃CN          | 5        | 70         | 0         | 39        |
| 24    | 0.5         | 3           | CH₃CN          | 2        | 20         | 0         | 67        |
| 25    | 0.5         | 3           | CH₂CN          | 2        | 40         | 0         | 87        |
| 26    | 0.5         | 3           | CH₂CN          | 2        | 60         | 0         | 86        |
| 27    | 0.5         | 3           | CH₂CN          | 2        | 80         | 0         | 81        |
| 28    | 0.5         | 3           | CH₂CN          | 2        | 90         | 5         | 79        |

[a] Reaction conditions: 1a (0.8 mmol), 2a (1.6 mmol), I₂ (0.5 mmol), H₂O₂ (3 mmol), CH₃CN (5 mL), 2 h, 40 °C, in an oil bath, opened tube, [b] isolated yield.
Table S2. Optimization for generating 3c

![Chemical Structures](image_url)

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$^{[a]}$ Reaction conditions: 1b (1 mmol), 2a (1.75 mmol), I$_2$ (10 mmol%), H$_2$O$_2$ (7 mmol), CH$_3$CN (5 mL), 1 h, 80 °C, in an oil bath, opened tube. $^{[b]}$ isolated yield.
Scheme S1. Control Experiment

For generating 3a: R = H, 1a:2a = 2:1, I2 (10 mmol%), 3 h, 60 °C
TEMPO (60 °C) 79% 3a (without TEMPO, 84% 3a)
BHT (60 °C) 71% 3a (without BHT, 84% 3a)

For generating 3b: R = I, 1a:2a = 1:2, I2 (0.5 mmol), 2 h, 40 °C
TEMPO (40 °C) 84% 3b (without TEMPO, 87% 3b)
BHT (40 °C) 72% 3b (without BHT, 87% 3b)

1a + 2a $\xrightarrow{\text{kl (20 mmol%), H}_2\text{O}_2 (3 mmol)}_{\text{CH}_3\text{CN}, 60 \degree \text{C}, 3 h}$ 3a (0%);
1a + 2a $\xrightarrow{\text{kl (20 mmol%), O}_2}$ 3a (0%);
1a + 2a $\xrightarrow{\text{kl (1 mmol), H}_2\text{O}_2 (3 mmol)}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 h}$ 3b (0%);
1a + 2a $\xrightarrow{\text{kl (1 mmol), O}_2 (3 mmol)}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 h}$ 3b (0%);

1a + 2a $\xrightarrow{\text{l}_2 (10 \text{ mmol%}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 60 \degree \text{C}, 3 \text{ h}}$ 3b (69%);
1a + 2a $\xrightarrow{\text{l}_2 (0.5 \text{ mmol}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (81%);
1a + 2a $\xrightarrow{\text{l}_2 (0.5 \text{ mmol}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (92%);
1a + 2a $\xrightarrow{\text{PhCOOH (1a:2a:H}^+=1:1:1)}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (96%);
1a + 2a $\xrightarrow{\text{l}_2 (0.5 \text{ mmol}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (96%);
1a + 2a $\xrightarrow{\text{l}_2 (0.5 \text{ mmol}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (96%);
1a + 2a $\xrightarrow{\text{l}_2 (0.5 \text{ mmol}, \text{H}_2\text{O}_2 (3 \text{ mmol})}_{\text{CH}_3\text{CN}, 40 \degree \text{C}, 2 \text{ h}}$ 3b (96%);
(e) $1a = 0.8 \text{ mmol, } 2a = 1.6 \text{ mmol}$

\[
\begin{align*}
1a+2a & \xrightarrow{I_2 (0.5 \text{ mmol}), O_2} \text{CH}_3\text{CN, 40 °C, 2 h} & 3b (0\%) \text{ and } 3a (71\%) \\
1a+2a & \xrightarrow{I_2 (0.5 \text{ mmol}), N_2} \text{CH}_3\text{CN, 40 °C, 2 h} & 3b (0\%) \text{ and } 3a (61\%)
\end{align*}
\]
3. General Procedure

1.1. General Procedure A
General Procedures for the Synthesis of 3-phenyl-imidazo[1,5-a]pyridine by benzaldehyde and 2-pyridinemethanamine. 1a (0.8 mmol, 2 eq.), 2a (0.4 mmol, 1 eq.), I2 (25.4 mg, 10 mmol%), 30% hydrogen peroxide (3 mmol), CH3CN (5 mL) were transferred into a 25 mL round-bottom flask equipped with a spherical condensing tube. Then the reaction flask was placed in a heating block that was preheated to 60 °C. The reaction was monitored by checking the TLC. After some time of 3 h, the reaction flask was allowed to cool to room temperature. The mixture was concentrated under reduced pressure and the residue was purified by flash chromatography on silica gel eluting with petroleum ether/EtOAc (v/v = 20:1 to 5:1) to afford the products 3a.

1.2. General Procedure B
General Procedures for the Synthesis of 1-iodo-3-phenyl-imidazo[1,5-a]pyridine, by benzaldehyde and 2-pyridinemethanamine. 1a (0.8 mmol, 1 eq.), 2a (1.6 mmol, 2 eq.), I2 (127 mg, 0.5 mmol), 30% hydrogen peroxide (3 mmol), CH3CN (5 mL) were transferred into a 25 mL round-bottom flask equipped with a spherical condensing tube. Then the reaction flask was placed in a heating block that was preheated to 40 °C. The reaction was monitored by checking the TLC. After some time of 2 h, the reaction flask was allowed to cool to room temperature. The mixture was concentrated under reduced pressure and the residue was purified by flash chromatography on silica gel eluting with petroleum ether/EtOAc (v/v = 15:1 to 3:1) to afford the products 3b.

1.3. General Procedure C
General Procedures for the Synthesis of 2-phenyl-1H-benzimidazole by benzaldehyde and 1, 2-benzenediamine. 1b (1 mmol, 1 eq.), 2a (1.75 mmol, 1.75 eq.), I2 (25.4 mg, 10 mmol%), 30% hydrogen peroxide (7 mmol), CH3CN (5 mL) were transferred into a 25 mL round-bottom flask equipped with a spherical condensing tube. Then the reaction flask was placed in a heating block that was preheated to 80 °C. The reaction was monitored by checking the TLC. After some time of 1 h, the reaction flask was allowed to cool to room temperature. The mixture was concentrated under reduced pressure and the residue was purified by flash chromatography on silica gel eluting with petroleum ether/EtOAc (v/v = 20:1 to 5:1) to afford the products 3c.
4. Photophysical Properties of Selected Products

![3an](image1.png) ![3aaa](image2.png) ![3aac](image3.png)

**Figure S1.** Partially selected emission of obtained 3an, 3aaa, and 3aac in four solvents under 365nm irradiation.

**Table S3.** Wavelengths of maximum absorbance and maximum emission and Stokes shift and the fluorescence quantum yields of selected products in DCM, Dioxane, MeOH, and DMSO.

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<th>λem(nm) (Stokes shift cm⁻¹)</th>
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Table S4 Fluorescence quantum yields of 20 selected compounds in DCM, Dioxane, MeOH and DMSO

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Figure S2. Partially electronic absorption and emission spectra of selected compounds recorded in DCM, Dioxane, MeOH, and DMSO (1 × 10^{-5} M).
**Table S5.** Comparison of experimental absorption wavelengths for selected compounds with theoretical calculations: electron transition, HOMO and LUMO energies, oscillator intensity, and absorption wavelength.

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5. Experimental Characterization Data for the Products

![Chemical Structure](image1)

3-Phenyl-imidazo[1, 5-a] pyridine (3a)\(^1\):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (65.2 mg, 84%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.10 (d, \(J = 6.8\) Hz, 1H), 7.66 (d, \(J = 6.76\) Hz, 2H), 7.49 - 7.16 (m, 5H), 6.55 (dd, \(J = 8.8, 6.0\) Hz, 1H), 6.38 (t, \(J = 6.8\) Hz, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 138.22, 131.61, 130.44, 128.96, 128.56, 127.89, 121.36, 120.63, 118.74, 113.03.

![Chemical Structure](image2)

3-(2-fluorophenyl)-imidazo[1,5-a]pyridine (3ab)\(^2\):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 2-fluoro-benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (72.9 mg, 86%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 7.80-7.60 (m, 2H), 7.52 (s, 1H), 7.36 (s, 2H), 7.45-7.31 (m, 1H), 7.14 (t, \(J = 9.2\) Hz, 1H), 6.66 (dd, \(J = 8.4, 5.88\) Hz, 1H), 6.48 (t, \(J = 6.8\) Hz, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 159.92 (d, \(J = 247.7\) Hz), 133.54, 132.22, 132.19, 131.78, 130.84 (d, \(J = 8.2\) Hz), 124.85 (d, \(J = 3.6\) Hz), 122.35, 122.29, 120.93, 119.03, 118.39, 116.18 (d, \(J = 21.5\) Hz), 112.80; \(^19\)F NMR (376 MHz, CDCl\(_3\)) \(\delta\): -111.14.

![Chemical Structure](image3)

3-(3-fluorophenyl)-imidazo[1,5-a]pyridine (3ac)\(^2\):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 3-fluoro-benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (73.8 mg, 87%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.06 (d, \(J = 7.4\) Hz, 1H), 7.45-7.34 (m, 3H), 7.32-7.27 (m, 2H), 6.94 (td, \(J = 8.6, 2.2\) Hz, 1H), 6.54 (dd, \(J = 8.9, 5.6\) Hz, 1H), 6.39 (t, \(J = 6.9\) Hz, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 162.90 (d, \(J = 247.5\) Hz), 136.78, 132.44, 131.90, 130.50 (d, \(J = 9.3\) Hz), 123.13 (d, \(J = 2.9\) Hz), 121.16, 120.90, 119.03, 118.74, 115.31 (d, \(J = 21.1\) Hz), 114.83, 114.60, 113.42; \(^19\)F NMR (376 MHz, CDCl\(_3\)) \(\delta\): -111.82.
3-(4-fluorophenyl)-imidazo[1,5-a]pyridine (3ad): From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 4-fluoro-benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (75.5 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.09 (d, $J = 7.2$ Hz, 1H), 7.78-7.61 (m, 2H), 7.46 (s, 1H), 7.40 (d, $J = 8.6$ Hz, 1H), 7.13 (t, $J = 8.6$ Hz, 2H), 6.64 (dd, $J = 8.8, 6.4$ Hz, 1H), 6.48 (t, $J = 6.9$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 162.91 (d, $J = 249.8$ Hz), 137.33, 131.59, 129.85 (d, $J = 8.3$ Hz), 126.66, 121.12, 120.59, 118.85, 118.76, 116.01 (d, $J = 21.6$ Hz), 113.22; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -112.10.

3-(2-chlorophenyl)-imidazo[1,5-a]pyridine (3ae): From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 2-chlorobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (76.6 mg, 84%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.05 (d, $J = 7.2$ Hz, 1H), 7.60 (d, $J = 8.5$ Hz, 2H), 7.43 (s, 1H), 7.35 (d, $J = 8.5$ Hz, 3H), 6.59 (dd, $J = 9.0, 6.4$ Hz, 1H), 6.43 (t, $J = 6.8$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 137.01, 134.31, 131.82, 129.18, 129.00, 128.91, 121.14, 120.88, 118.92, 118.82, 113.37.

3-(3-chlorophenyl)-imidazo[1,5-a]pyridine (3af): From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 3-chlorobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (77.5 mg, 85%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.04 (d, $J = 7.0$ Hz, 1H), 7.64 (s, 1H), 7.50 (d, $J = 7.2$ Hz, 1H), 7.39 (s, 1H), 7.34-7.16 (m, 3H), 6.55 (t, $J = 6.6$ Hz, 1H), 6.39 (t, $J = 6.5$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 135.45, 133.77, 131.02, 130.82, 129.06, 127.30, 126.65, 124.45, 120.02, 119.89, 117.99, 117.64, 112.38.
3-(4-chlorophenyl)-imidazo[1,5-a]pyridine (3ag)\(^{[1]}\):

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 4-chlorobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (76.6 mg, 84\%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.10 (d, \(J = 7.4\) Hz, 1H), 7.85-7.65 (m, 2H), 7.46 (s, 1H), 7.41 (d, \(J = 9.1\) Hz, 1H), 7.14 (t, \(J = 8.6\) Hz, 2H), 6.66 (dd, \(J = 9.8, 6.3\) Hz, 1H), 6.49 (t, \(J = 6.6\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 134.56, 133.06, 131.87, 129.98, 129.49, 128.82, 128.32, 125.98, 120.99, 119.05, 117.83, 117.23, 111.45.

3-(2-bromophenyl)-imidazo[1,5-a]pyridine (3ah)\(^{[3]}\):

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 2-bromobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (91.4 mg, 84\%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 7.61 (d, \(J = 7.4\) Hz, 1H), 7.51-7.42 (m, 3H), 7.39 (d, \(J = 8.6\) Hz, 1H), 7.34 (t, \(J = 8.0\) Hz, 1H), 7.29-7.20 (m, 1H), 6.64 (dd, \(J = 9.1, 6.8\) Hz, 1H), 6.44 (t, \(J = 5.6\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 136.89, 133.26, 133.18, 131.58, 130.93, 127.69, 124.20, 122.12, 119.97, 119.02, 118.41, 112.62.

3-(3-bromophenyl)-imidazo[1,5-a]pyridine (3ai)\(^{[3]}\):

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 3-bromobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (92.4 mg, 85\%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.08 (d, \(J = 7.1\) Hz, 1H), 7.83 (s, 1H), 7.58 (d, \(J = 7.8\) Hz, 1H), 7.45-7.37 (m, 2H), 7.34 (d, \(J = 8.4\) Hz, 1H), 7.22 (t, \(J = 7.6\) Hz, 1H), 6.60 (dd, \(J = 8.9, 6.4\) Hz, 1H), 6.44 (t, \(J = 7.6\) 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 136.49, 132.37, 131.94, 131.39, 130.69, 130.42, 126.09, 123.04, 121.15, 121.02, 119.13, 118.81, 113.54.
3-(4-bromophenyl)-imidazo[1,5-a]pyridine (3a)

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 4-bromobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (92.4 mg, 85%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.03 (d, $J$ = 6.6 Hz, 1H), 7.58-7.40 (m, 4H), 7.41 (s, 1H), 7.32 (d, $J$ = 9.5 Hz, 1H), 6.57 (dd, $J$ = 8.8, 6.3 Hz, 1H), 6.41 (t, $J$ = 6.7 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 136.99, 136.43, 132.11, 131.85, 129.33, 129.20, 122.48, 121.13, 120.93, 118.97, 118.82, 113.43.

3-(2-nitrophenyl)-imidazo[1,5-a]pyridine (3ak)

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 2-nitrobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (85.1 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.14 (d, $J$ = 7.7 Hz, 1H), 7.80-7.70 (m, 2H), 7.67-7.57 (m, 3H), 7.52 (d, $J$ = 7.7 Hz, 1H), 6.77 (dd, $J$ = 9.2, 6.6 Hz, 1H), 6.56 (t, $J$ = 6.6 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 148.67, 133.43, 133.29, 132.84, 131.65, 130.18, 125.07, 125.04, 120.96, 120.89, 119.40, 118.74, 113.53.

3-(3-nitrophenyl)-imidazo[1,5-a]pyridine (3al)

From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 3-nitrobenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (88.0 mg, 92%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.70 (s, 1H), 8.36-8.25 (m, 2H), 8.21 (d, $J$ = 8.0 Hz, 1H), 7.72 (t, $J$ = 8.0 Hz, 1H), 7.63 (s, 1H), 7.57 (d, $J$ = 8.0 Hz, 1H), 6.84 (dd, $J$ = 8.6, 6.1 Hz, 1H), 6.72 (t, $J$ = 7.0 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 148.65, 135.63, 133.64, 132.44, 132.15, 130.14, 122.95, 121.97, 121.63, 120.92, 119.60, 119.08, 114.22.
3-(4-nitrophenyl)-imidazo[1,5-a]pyridine (3am): 
From 2-pyridinmethanamine (0.8 mmol, 2 eq.) and 4-nitro-benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (89.9 mg, 94%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.25 (d, $J = 8.2$ Hz, 3H), 7.91 (d, $J = 8.3$ Hz, 2H), 7.54 (s, 1H), 7.46 (d, $J = 8.9$ Hz, 1H), 6.75 (dd, $J = 9.2$, 6.4 Hz, 1H), 6.62 (t, $J = 6.6$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 146.91, 136.50, 135.80, 132.93, 132.93, 127.68, 124.36, 122.35, 121.29, 120.05, 119.13, 114.47.

4-imidazo[1,5-a]pyridin-3-yl-benzonitrile (3an): 
From 2-pyridinmethanamine (0.8 mmol, 2 eq.) and 4-formylbenzonitrile (0.4 mmol, 1 eq.), following the general procedure A, the title compound (82.3 mg, 94%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.22 (d, $J = 7.2$ Hz, 1H), 7.88 (d, $J = 8.1$ Hz, 2H), 7.71 (d, $J = 8.5$ Hz, 2H), 7.54 (s, 1H), 7.47 (d, $J = 8.4$ Hz, 1H), 6.75 (dd, $J = 9.0$, 6.4 Hz, 1H), 6.60 (t, $J = 6.8$, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 136.11, 134.71, 132.77, 132.66, 127.75, 124.00, 121.19, 119.78, 119.10, 118.66, 114.21, 111.49.

3-(4-(trifluoromethyl)phenyl)imidazo[1,5-a]pyridine (3ao): 
From 2-pyridinmethanamine (0.8 mmol, 2 eq.) and 4-(trifluoromethyl)benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (102.7 mg, 98%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.18 (d, $J = 6.8$ Hz, 1H), 7.84 (d, $J = 8.2$ Hz, 2H), 7.67 (d, $J = 8.2$ Hz, 2H), 7.50 (s, 1H), 7.43 (d, $J = 9.1$ Hz, 1H), 6.69 (dd, $J = 8.8$, 6.4 Hz, 1H), 6.54 (t, $J = 7.0$, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 136.69, 133.90, 132.25, 130.22 (d, $J = 32.8$ Hz), 127.87, 126.00 (q, $J = 3.7$ Hz), 124.01 (d, $J = 272.7$ Hz), 121.41, 121.17, 119.35, 119.00, 113.78; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -62.65.

3-(2-methylphenyl)-imidazo[1,5-a]pyridine (3ap): 
From 2-pyridinmethanamine (0.8 mmol, 2 eq.) and 2-methylbenzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (54.1 mg, 65%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.18 (d, $J = 6.8$ Hz, 1H), 7.84 (d, $J = 8.2$ Hz, 2H), 7.67 (d, $J = 8.2$ Hz, 2H), 7.50 (s, 1H), 7.43 (d, $J = 9.1$ Hz, 1H), 6.69 (dd, $J = 8.8$, 6.4 Hz, 1H), 6.54 (t, $J = 7.0$, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 136.69, 133.90, 132.25, 130.22 (d, $J = 32.8$ Hz), 127.87, 126.00 (q, $J = 3.7$ Hz), 124.01 (d, $J = 272.7$ Hz), 121.41, 121.17, 119.35, 119.00, 113.78; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -62.65.
white solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 7.64 (d, $J$ = 7.1 Hz, 1H), 7.58 (s, 1H), 7.48 (dd, $J$ = 12.5, 9.4 Hz, 2H), 7.40-7.28 (m, 3H), 6.72 (dd, $J$ = 9.2, 6.4 Hz, 1H), 6.50 (t, $J$ = 6.7 Hz, 1H), 2.24 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 138.41, 137.87, 130.83, 130.54, 130.38, 129.48, 129.36, 126.00, 121.49, 119.76, 118.56, 118.52, 112.59, 19.76.

3-(3-methylphenyl)-imidazo[1,5-a]pyridine (3aq)

3-(4-methylphenyl)-imidazo[1,5-a]pyridine (3ar)

3-(2-methoxyphenyl)-imidazo[1,5-a]pyridine (3as)
(t, J = 6.7 Hz, 1H), 3.78 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 157.30, 136.22, 132.46, 131.27, 130.63, 123.13, 121.10, 120.13, 119.42, 118.50, 118.17, 111.77, 111.22, 55.51.

3-(2-methoxyphenyl)-imidazo[1, 5-a]pyridine (3at)$^{[2]}$:
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 4-methoxy-benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (63.6 mg, 71%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.06 (d, J = 7.3 Hz, 1H), 7.61 (d, J = 7.5 Hz, 2H), 7.42 (s, 1H), 7.34 (d, J = 9.2 Hz, 1H), 6.94 (d, J = 6.7 Hz, 2H), 6.57 (dd, J = 9.2, 6.3 Hz, 1H), 6.40 (t, J = 6.8 Hz, 1H), 3.76 (s, 3H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 159.84, 138.22, 131.28, 129.37, 122.92, 121.33, 120.20, 118.73, 118.43, 114.39, 112.82, 55.37.

3-(naphthalen-2-yl)imidazo[1,5-a]pyridine (3au)$^{[3]}$:
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and 2-naphthaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (71.2 mg, 73%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.41 (d, J = 7.3 Hz, 1H), 8.29 (s, 1H), 8.04-7.91 (m, 4H), 7.64 (s, 1H), 7.58-7.52 (m, 3H), 6.82-6.73 (m, 1H), 6.62 (t, J = 6.8 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 138.28, 133.37, 133.15, 131.83, 128.81, 128.24, 127.85, 126.69, 126.66, 126.63, 125.72, 121.51, 120.96, 118.91, 113.25.

3-(thiophen-2-yl)imidazo[1,5-a]pyridine (3av)$^{[1]}$:
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and thiophene-2-carbaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (56.8 mg, 71%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.21 (d, J = 7.2 Hz, 1H), 7.47-7.36 (m, 3H), 7.31 (d, J = 5.2 Hz, 1H), 7.08 (dd, J = 4.8, 3.6 Hz, 1H), 6.64 (dd, J = 9.2, 6.4 Hz, 1H), 6.54 (t, J = 7.2, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 133.03, 132.73, 131.72, 127.62, 125.84, 124.30, 121.68, 120.93, 118.80, 118.79, 113.67.
3-(furan-2-yl)imidazo[1,5-a]pyridine (3aw)\(^1\):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and furan-2-carbaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (47.8 mg, 65%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.71 (s, 1H), 7.38 (s, 1H), 7.09 (s, 1H), 6.52 (s, 1H), 6.42-6.21 (m, 1H), 5.94 (s, 1H), 5.61 (s, 1H), 5.48 (s, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 151.11, 148.14, 136.53, 132.93, 126.04, 121.73, 121.58, 120.94, 120.16, 117.99, 113.55.

4-(imidazo[1,5-a]pyridin-3-yl)thiazole (3ax):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and furan-thiazole-4-carbaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (55.5 mg, 69%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). m.p.: 95-97 °C; IR (KBr) 3116, 1629, 1541, 1502, 1458, 1344, 1308, 1253, 1134, 1097, 1002, 881, 810, 746, 694, 655 cm\(^{-1}\); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 9.37 (d, \(J = 7.3\) Hz, 1H), 8.85 (s, 1H), 7.99 (s, 1H), 7.50-7.34 (m, 2H), 6.71 (dd, \(J = 9.1, 6.3\) Hz, 1H), 6.59 (t, \(J = 6.8\) Hz, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 152.62, 148.39, 132.93, 131.56, 124.72, 120.58, 119.36, 118.11, 115.39, 113.20; HRMS(ESI): calculated for C\(_{10}\)H\(_8\)N\(_3\)S\(^+\) [M+H]\(^+\): 202.0433, found: 202.0434.

3-(pyridin-2-yl)imidazo[1,5-a]pyridine (3ay)\(^1\):
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and picolinaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (60.8 mg, 78%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 9.84 (d, \(J = 7.2\) Hz, 1H), 8.51 (d, \(J = 5.0\) Hz, 1H), 8.23 (d, \(J = 8.1\) Hz, 1H), 7.65 (t, \(J = 7.8\) Hz, 1H), 7.48 (s, 1H), 7.40 (d, \(J = 9.0\) Hz, 1H), 7.09-7.00 (m, 1H), 6.72 (dd, \(J = 9.1, 6.4\) Hz, 1H), 6.59 (t, \(J = 7.2\) Hz, 1H); \(^13\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 151.15, 148.09, 136.45, 135.41, 132.91, 126.01, 121.64, 121.49, 121.02, 120.07, 117.92, 113.45.
3-(pyridin-3-yl)imidazo[1,5-a]pyridine (3az)[3]:
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and nicotinaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (59.3 mg, 76%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.99 (s, 1H), 8.56 (s, 1H), 8.14 (d, $J = 7.2$ Hz, 1H), 8.02 (d, $J = 7.9$ Hz, 1H), 7.50 (s, 1H), 7.41 (d, $J = 9.1$ Hz, 1H), 7.35 (dd, $J = 8.0$, 4.8 Hz, 1H), 6.67 (dd, $J = 9.2$, 6.4 Hz, 1H), 6.52 (t, $J = 6.8$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 149.34, 148.47, 135.16, 132.12, 126.78, 123.77, 121.40, 120.95, 119.29, 118.91, 113.76.

3-(pyridin-4-yl)imidazo[1,5-a]pyridine (3aaa)[3]:
From 2-pyridinemethanamine (0.8 mmol, 2 eq.) and isonicotinaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (64.0 mg, 82%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.64 (d, $J = 4.3$ Hz, 2H), 8.28 (d, $J = 7.2$ Hz, 1H), 7.66 (d, $J = 4.4$ Hz, 2H), 7.53 (s, 1H), 7.45 (d, $J = 9.1$ Hz, 1H), 6.74 (dd, $J = 9.1$, 6.4 Hz, 1H), 6.61 (t, $J = 6.8$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 150.41, 137.67, 135.18, 134.50, 132.84, 121.99, 121.41, 121.19, 119.87, 119.06, 114.26.

3-phenyl-1-(pyridin-2-yl)imidazo[1,5-a]pyridine (3aab)[1]:
From di(pyridin-2-yl)methanamine (0.8 mmol, 2 eq.) and benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (74.8 mg, 69%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.61 (d, $J = 9.2$ Hz, 1H), 8.54 (d, $J = 4.9$ Hz, 1H), 8.15 (t, $J = 7.9$ Hz, 2H), 7.74 (d, $J = 8.0$ Hz, 2H), 7.62 (t, $J = 7.8$ Hz, 1H), 7.45 (t, $J = 7.5$ Hz, 2H), 7.36 (t, $J = 7.4$ Hz, 1H), 7.04-6.95 (m, 1H), 6.81 (dd, $J = 9.3$, 6.3 Hz, 1H), 6.53 (t, $J = 6.8$ Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 155.06, 148.99, 138.07, 136.26, 130.58, 130.23, 130.14, 129.07, 128.94, 128.40, 121.85, 121.61, 121.06, 120.47, 119.96, 113.93.
1,3-diphenylimidazo[1,5,a]pyridine (3aac)\textsuperscript{[1]}:
From phenyl(pyridin-2-yl)methanamine (0.8 mmol, 2 eq.) and benzaldehyde (0.4 mmol, 1 eq.), following the general procedure A, the title compound (82.1 mg, 76%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \( ^1\)H NMR (400 MHz, CDCl\textsubscript{3}) \( \delta \): 8.16 (d, \( J = 7.2 \) Hz, 1H), 7.87 (d, \( J = 8.0 \) Hz, 2H), 7.77 (d, \( J = 8.0 \) Hz, 3H), 7.50-7.43 (m, 2H), 7.42-7.34 (m, 3H), 7.26-7.19 (m, 1H), 6.71 (dd, \( J = 9.3, 6.3 \) Hz, 1H), 6.50 (t, \( J = 6.8 \) Hz, 1H); \( ^{13}\)C NMR (101 MHz, CDCl\textsubscript{3}) \( \delta \): 138.14, 134.99, 132.04, 130.21, 129.04, 128.83, 128.73, 128.35, 127.69, 126.83, 126.55, 121.79, 119.70, 119.19, 113.24.

1-iodo-3-phenylimidazo[1,5,a]pyridine (3b)\textsuperscript{[4]}:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and benzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (222.7 mg, 87%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). \( ^1\)H NMR (400 MHz, CDCl\textsubscript{3}) \( \delta \): 7.95 (d, \( J = 7.2 \) Hz, 1H), 7.54 (d, \( J = 7.6 \) Hz, 2H), 7.36-7.18 (m, 3H), 7.10 (d, \( J = 9.2 \) Hz, 1H), 6.54 (dd, \( J = 9.2, 6.4 \) Hz, 1H), 6.34 (t, \( J = 6.8 \) Hz, 1H); \( ^{13}\)C NMR (101 MHz, CDCl\textsubscript{3}) \( \delta \): 140.30, 133.32, 129.28, 129.01, 127.84, 121.73, 120.31, 118.76, 114.01, 74.27.

3-(2-fluorophenyl)-1-iodoimidazo[1,5,a]pyridine (3bb):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-fluorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (235.2 mg, 87%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 122-124 °C; IR (KBr) 3053, 1627, 1568, 1510, 1454, 1355, 1307, 1259, 1209, 1091, 1002, 943, 815, 765, 732, 684 cm\textsuperscript{-1}; \( ^1\)H NMR (400 MHz, CDCl\textsubscript{3}) \( \delta \): 7.77-7.67 (m, 2H), 7.45 (tdd, \( J = 7.5, 5.2, 1.8 \) Hz, 1H), 7.34 (d, \( J = 9.6 \) Hz, 1H), 7.29 (t, \( J = 7.6 \) Hz, 1H), 7.25-7.15 (m, 1H), 6.81 (dd, \( J = 9.2, 6.4 \) Hz, 1H), 6.61 (t, \( J = 6.8 \) Hz, 1H); \( ^{13}\)C NMR (101 MHz, CDCl\textsubscript{3}) \( \delta \): 159.75 (d, \( J = 249.7 \) Hz), 135.68, 133.69, 132.40, 132.36, 131.34 (d, \( J = 8.3 \) Hz), 124.96 (d, \( J = 3.4 \) Hz), 122.86, 122.79, 120.54, 118.53,
117.36, 117.22, 116.12 (d, J = 21.4 Hz), 113.74, 74.09; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -110.71; HRMS(ESI): calculated for C$_{13}$H$_9$FIN$_2$ $[M+H]^+$: 338.9789, found: 338.9797.

3-(3-fluorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bc):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 3-fluorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (238.0 mg, 88%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 66–68 °C; IR (KBr) 3076, 1612, 1583, 1568, 1419, 1442, 1116, 1018, 862, 788, 738, 692 cm$^{-1}$; $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.24 (d, J = 7.2 Hz, 1H), 7.57 (d, J = 7.8 Hz, 1H), 7.54–7.45 (m, 2H), 7.38 (d, J = 9.2 Hz, 1H), 7.19–7.09 (m, 1H), 6.84 (dd, J = 9.2, 6.4 Hz, 1H), 6.67 (t, J = 6.8 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 163.00 (d, J = 247.3 Hz), 139.10, 133.73, 131.39, 131.30, 130.70 (d, J = 8.5 Hz), 123.32 (d, J = 3.0 Hz), 121.70, 120.53, 119.13, 116.09, 115.88, 114.90 (d, J = 23.1 Hz), 114.40, 74.55; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -111.49; HRMS(ESI): calculated for C$_{13}$H$_9$FIN$_2$ $[M+H]^+$: 338.9789, found: 338.9791.

3-(4-fluorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bd)$^{[4]}$:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-fluorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (240.7 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). $^1$H NMR (400 MHz, CDCl$_3$) δ: 8.01 (d, J = 7.2 Hz, 1H), 7.67–7.56 (m, 2H), 7.22 (d, J = 9.2 Hz, 1H), 7.08 (t, J = 8.7 Hz, 2H), 6.67 (dd, J = 9.2, 6.4 Hz, 1H), 6.49 (t, J = 6.8 Hz, 1H); $^{13}$C NMR (101 MHz, CDCl$_3$) δ: 162.96 (d, J = 249.7 Hz), 139.44, 133.36, 129.90 (d, J = 8.4 Hz), 125.53 (d, J = 3.4 Hz), 121.54, 120.28, 118.97, 116.20 (d, J = 21.9 Hz), 114.16, 74.06; $^{19}$F NMR (376 MHz, CDCl$_3$) δ: -111.06.

3-(2-chlorophenyl)-1-iodoimidazo[1,5-a]pyridine (3be):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-chlorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (252.0 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 145-146 °C; IR (KBr)
3-(3-chlorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bf):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 3-chlorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (254.9 mg, 90%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 103-105 °C; IR (KBr) 3055, 1593, 1558, 1496, 1452, 1353, 1301, 1257, 1093, 1006, 947, 887, 786, 752, 690, 563 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 7.65-7.56 (m, 2H), 7.57-7.51 (m, 1H), 7.50-7.35 (m, 3H), 6.87 (dd, J = 9.2, 6.5 Hz, 1H), 6.64 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 137.95, 134.05, 133.37, 133.19, 131.10, 129.98, 128.52, 127.26, 122.78, 120.48, 118.63, 113.54, 73.27; HRMS(ESI): calculated for C₁₃H₉ClIN₂⁺ [M+H]⁺: 354.9493, found: 354.9495.

![3-(3-chlorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bf)](image)

3-(4-chlorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bg)⁴:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-chlorobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (257.7 mg, 91%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 103-105 °C; IR (KBr) 3055, 1593, 1558, 1496, 1452, 1353, 1301, 1257, 1093, 1006, 947, 887, 786, 752, 690, 563 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.21 (d, J = 7.1 Hz, 1H), 7.78 (s, 1H), 7.66 (dt, J = 7.4, 1.6 Hz, 1H), 7.48-7.34 (m, 3H), 6.84 (dd, J = 9.2, 6.4 Hz, 1H), 6.67 (t, J = 6.8, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 138.88, 135.05, 133.73, 131.02, 129.03, 127.89, 121.65, 120.59, 119.09, 114.47, 74.64; HRMS(ESI): calculated for C₁₃H₉ClIN₂⁺ [M+H]⁺: 354.9493, found: 354.9493.

![3-(4-chlorophenyl)-1-iodoimidazo[1,5-a]pyridine (3bg)](image)

3-(2-bromophenyl)-1-iodoimidazo[1,5-a]pyridine (3bh):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-bromobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (289.7 mg, 91%) was obtained as a faint
yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 122-125 °C; IR (KBr) 3070, 1627, 1554, 1496, 1357, 1259, 1114, 1002, 945, 740, 686 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 7.73 (d, J = 8.0 Hz, 1H), 7.64-7.54 (m, 2H), 7.47 (t, J = 7.5 Hz, 1H), 7.44-7.35 (m, 2H), 6.88 (dd, J = 9.2, 6.5 Hz, 1H), 6.65 (t, J = 6.8, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 139.09, 133.62, 133.16, 132.98, 131.33, 130.61, 127.77, 123.94, 122.76, 120.48, 118.64, 113.51, 73.01; HRMS(ESI): calculated for C₁₃H₉BrIN₂⁺ [M+H]⁺: 398.8988, found: 398.8993.

3-(3-bromophenyl)-1-iodoimidazo[1,5-a]pyridine (3bi):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 3-bromobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (302.5 mg, 95%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 107-109 °C; IR (KBr) 3078, 1631, 1593, 1556, 1500, 1421, 1352, 1299, 1257, 1068, 1010, 950, 879, 740, 686 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.21 (d, J = 7.2 Hz, 1H), 7.95 (s, 1H), 7.70 (d, J = 7.7 Hz, 1H), 7.56 (d, J = 8.0 Hz, 1H), 7.45-7.32 (m, 2H), 6.84 (dd, J = 9.2, 6.4 Hz, 1H), 6.67 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 138.76, 133.75, 131.96, 131.27, 130.79, 130.49, 126.15, 123.14, 121.64, 120.60, 119.13, 114.48, 74.65; HRMS(ESI): calculated for C₁₃H₉BrIN₂⁺ [M+H]⁺: 398.8988, found: 398.8987.

3-(4-bromophenyl)-1-iodoimidazo[1,5-a]pyridine (3bj) [⁴]:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-bromobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (305.7 mg, 96%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.08 (d, J = 7.2 Hz, 1H), 7.55 (s, 4H), 7.28 (d, J = 9.2 Hz, 1H), 6.74 (dd, J = 9.2, 6.4 Hz, 1H), 6.56 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 139.32, 133.76, 131.96, 131.27, 130.79, 130.49, 126.15, 123.13, 121.62, 120.43, 119.16, 114.36, 74.52.

1-iodo-3-(2-nitrophenyl)imidazo[1,5-a]pyridine (3bk):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-nitrobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (262.8 mg, 90%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 158-161 °C; IR (KBr) 3095, 1612, 1516, 1340, 1257, 1137, 1002, 945, 854, 786, 744, 698 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.10 (t, J = 8.8 Hz, 1H), 7.72-7.55 (m, 3H), 7.44 (d, J = 7.2 Hz, 1H), 7.30 (t, J = 9.1 Hz, 1H), 6.78 (t, J = 7.2 Hz, 1H), 6.54 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 148.25, 135.61, 133.80, 133.57, 133.52, 130.80, 125.18, 124.10, 121.51, 120.87, 118.90, 114.41, 73.66; HRMS(ESI): calculated for C₁₃H₉IN₃O₂⁺ [M+H]⁺: 365.9734, found: 365.9730.

1-iodo-3-(3-nitrophenyl)imidazo[1,5-a]pyridine (3bl):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 3-nitrobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (277.4 mg, 95%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 200-202 °C; IR (KBr) 3086, 1705, 1595, 1344, 1244, 1103, 1108, 1006, 854, 812, 738, 692 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.58 (s, 1H), 8.20 (d, J = 8.6 Hz, 2H), 8.10 (d, J = 7.8 Hz, 1H), 7.64 (t, J = 8.0 Hz, 1H), 7.37 (d, J = 9.2 Hz, 1H), 6.84 (dd, J = 9.2, 6.4 Hz, 1H), 6.69 (t, J = 6.9 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 148.60, 137.80, 134.23, 133.72, 131.06, 130.25, 123.42, 121.95, 121.36, 121.03, 119.38, 115.14, 75.30; HRMS(ESI): calculated for C₁₃H₉IN₃O₂⁺ [M+H]⁺: 365.9734, found: 365.9733.

1-iodo-3-(4-nitrophenyl)imidazo[1,5-a]pyridine (3bm):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-nitrobenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (286.2 mg, 98%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 129-130 °C; IR (KBr) 3084, 1705, 1595, 1344, 1244, 1103, 1108, 1006, 854, 812, 738, 692 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.23 (s, 3H), 7.88 (d, J = 8.6 Hz, 2H), 7.33 (d, J = 8.8 Hz, 1H), 6.84 (t, J = 7.6 Hz, 1H), 6.69 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 147.19, 137.97, 135.38, 134.67, 127.78, 124.38, 121.73, 121.47, 119.39, 115.38, 115.30, 76.30; HRMS(ESI): calculated for C₁₃H₉IN₃O₂⁺ [M+H]⁺: 365.9734, found: 365.9737.
4-(1-iodoimidazo[1,5-a]pyridin-3-yl)benzonitrile (3bn):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-formylbenzonitrile (1.6 mmol, 2 eq.), following the general procedure B, the title compound (262.2 mg, 95%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 167-169 °C; IR (KBr) 2221, 1606, 1494, 1359, 1263, 1120, 1010, 844, 740, 682 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.27 (d, J = 7.2 Hz, 1H), 7.91 (d, J = 8.0 Hz, 2H), 7.77 (d, J = 8.0 Hz, 2H), 7.41 (d, J = 9.1 Hz, 1H), 6.90 (dd, J = 9.2, 6.4 Hz, 1H), 6.74 (t, J = 6.8, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 138.25, 134.40, 133.51, 132.79, 127.76, 121.66, 121.24, 119.33, 118.49, 115.14, 111.93, 75.86; HRMS(ESI): calculated for C₁₄H₉IN₃⁺ [M+H]⁺: 345.9836, found: 345.9837.

1-iodo-3-(4-(trifluoromethyl)phenyl)imidazo[1,5-a]pyridine (3bo) [⁴]:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-(trifluoromethyl)benzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (304.2 mg, 98%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.09 (d, J = 7.3 Hz, 1H), 7.75 (d, J = 8.2 Hz, 2H), 7.61 (d, J = 8.2 Hz, 2H), 7.23 (d, J = 9.2 Hz, 1H), 6.71 (dd, J = 9.3, 6.4 Hz, 1H), 6.54 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 138.74, 133.98, 132.72, 130.48 (d, J = 32.6 Hz), 127.80, 125.94 (q, J = 3.8 Hz), 123.90 (d, J = 272.3 Hz), 121.56, 120.83, 119.10, 114.71, 75.11; ¹⁹F NMR (376 MHz, CDCl₃) δ: -62.61.

1-iodo-3-(2-methylphenyl)imidazo[1,5-a]pyridine (3bp) [⁴]:
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-methylbenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (197.7 mg, 74%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). ¹H NMR (400 MHz, CDCl₃) δ: 7.59 (d, J = 7.2 Hz, 1H), 7.48-7.21 (m, 5H), 6.78 (dd, J = 9.2, 6.4 Hz, 1H), 6.54 (t, J = 6.7 Hz, 1H), 2.20 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ: 140.05, 138.34, 132.53, 130.86, 130.48, 129.80, 128.46, 126.09, 121.94, 120.10, 118.67, 113.58, 72.89, 19.78.
1-iodo-3-(3-methylphenyl)imidazo[1,5-a]pyridine (3bq):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 3-methylbenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (211.1 mg, 79%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 91-94 °C; IR (KBr) 2922, 1604, 1506, 1469, 1352, 1307, 1257, 1018, 948, 802, 751, 704 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.17 (d, J = 7.2 Hz, 1H), 7.57 (s, 1H), 7.51 (d, J = 7.7 Hz, 1H), 7.36 (t, J = 7.7 Hz, 1H), 7.30 (d, J = 9.2 Hz, 1H), 7.22 (d, J = 7.6 Hz, 1H), 6.74 (dd, J = 9.2, 6.4 Hz, 1H), 6.54 (t, J = 6.8 Hz, 1H), 2.41 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ: 140.59, 138.91, 133.30, 129.86, 129.21, 128.82, 128.79, 124.63, 121.90, 120.18, 118.86, 113.88, 74.08, 21.52; HRMS(ESI): calculated for C₁₄H₁₂IN₂⁺ [M+H]⁺: 335.0040, found: 335.0037.

1-iodo-3-(4-methylphenyl)imidazo[1,5-a]pyridine (3br):[4]
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-methylbenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (219.1 mg, 82%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.20 (d, J = 7.2 Hz, 1H), 7.66 (d, J = 6.4 Hz, 2H), 7.46-7.30 (m, 3H), 6.79 (dd, J = 9.1, 6.3 Hz, 1H), 6.59 (t, J = 6.8 Hz, 1H), 2.44 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ: 140.66, 139.14, 133.24, 129.69, 127.91, 126.50, 121.90, 120.03, 118.98, 113.77, 73.89, 21.45.

1-iodo-3-(2-methoxyphenyl)imidazo[1,5-a]pyridine (3bs):[4]
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-methoxybenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (196.0 mg, 70%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). ¹H NMR (400 MHz, CDCl₃) δ: 7.58-7.45 (m, 2H), 7.39 (t, J = 7.9 Hz, 1H), 7.28 (d, J = 9.1 Hz, 1H), 7.02 (t, J = 7.5 Hz, 1H), 6.95 (d, J = 8.3 Hz, 1H), 6.73 (dd, J = 9.2, 6.4 Hz, 1H), 6.48 (t, J = 6.8 Hz, 1H), 3.72 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ: 157.15, 138.47, 133.23, 132.74, 131.09, 123.71, 121.20, 119.97, 118.39, 112.66, 111.14, 73.09, 55.57.
1-iodo-3-(4-methoxyphenyl)imidazo[1,5-a]pyridine (3bt)\([4]\):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 4-methoxybenzaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (210.0 mg, 75%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.03 (d, \(J = 7.2\) Hz, 1H), 7.62–7.51 (m, 2H), 7.21 (d, \(J = 9.2\) Hz, 1H), 6.95–6.90 (m, 2H), 6.66 (dd, \(J = 9.2, 6.4\) Hz, 1H), 6.47 (t, \(J = 6.8\) Hz, 1H), 3.76 (s, 3H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 160.21, 140.49, 133.08, 130.05, 129.49, 121.81, 121.74, 119.98, 118.94, 114.47, 114.23, 113.78, 73.52, 55.44.

1-iodo-3-(naphthalen-2-yl)imidazo[1,5-a]pyridine (3bu):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and 2-naphthaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (233.8 mg, 79%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 120–121 °C; IR (KBr) 3055, 1628, 1598, 1498, 1438, 1363, 1265, 1199, 1112, 1008, 954, 860, 825, 744, 684 cm\(^{-1}\); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.21 (d, \(J = 7.2\) Hz, 1H), 8.10 (s, 1H), 7.85 (d, \(J = 8.5\) Hz, 1H), 7.81–7.73 (m, 3H), 7.46–7.36 (m, 2H), 7.27 (d, \(J = 9.2\) Hz, 1H), 6.70 (dd, \(J = 9.2, 6.4\) Hz, 1H), 6.51 (t, \(J = 6.8\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 140.48, 133.58, 133.29, 133.24, 128.87, 128.30, 127.86, 126.95, 126.88, 126.78, 126.70, 125.43, 121.90, 120.38, 119.08, 114.15, 74.50; HRMS(ESI): calculated for C\(_{17}\)H\(_{12}\)IN\(_2\)\([\text{M+H]}^+\): 371.0040, found: 371.0035.

1-iodo-3-(thiophen-2-yl)imidazo[1,5-a]pyridine (3bv)\([5]\):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and thiophene-2-carbaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (200.8 mg, 77%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.21 (d, \(J = 7.2\) Hz, 1H), 7.44 (d, \(J = 3.7\) Hz, 1H), 7.35 (d, \(J = 5.1\) Hz, 1H), 7.29 (d, \(J = 3.7\) Hz, 1H); 7.23 (d, \(J = 5.1\) Hz, 1H), 7.17–7.10 (m, 2H), 6.69 (d, \(J = 9.2\) Hz, 1H), 6.63 (dd, \(J = 9.2, 6.4\) Hz, 1H), 6.50 (t, \(J = 6.8\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 140.48, 133.58, 133.29, 133.24, 128.87, 128.30, 127.86, 126.95, 126.88, 126.78, 126.70, 125.43, 121.90, 120.38, 119.08, 114.15, 74.50; HRMS(ESI): calculated for C\(_{17}\)H\(_{12}\)N\(_2\)\([\text{M+H]}^+\): 371.0040, found: 371.0035.
3-(furan-2-yl)-1-iodomidazo[1,5-a]pyridine (3bw):

From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and furan-2-carbaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (186.0 mg, 75%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 8.33 (d, \(J = 7.2\) Hz, 1H), 7.55 (d, \(J = 3.7\) Hz, 1H), 7.46 (d, \(J = 5.1\) Hz, 1H), 7.40 (d, \(J = 9.2\) Hz, 1H), 7.20 (dd, \(J = 5.1, 3.7\) Hz, 1H), 6.86 (dd, \(J = 9.2, 6.4\) Hz, 1H), 6.73 (t, \(J = 6.8\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 147.19, 137.97, 135.30, 134.67, 127.78, 124.38, 121.73, 121.47, 119.39, 115.38, 76.30.

4-(1-iodoimidazo[1,5-a]pyridin-3-yl)thiazole (3bx):

From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and thiazole-4-carbaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (211.9 mg, 81%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 117–119 °C; IR (KBr) 3074, 1625, 1500, 1452, 1342, 1303, 1257, 1103, 995, 956, 883, 813, 740, 686 cm\(^{-1}\); \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 9.44 (d, \(J = 7.3\) Hz, 1H), 8.93 (d, \(J = 2.2\) Hz, 1H), 8.12 (d, \(J = 2.2\) Hz, 1H), 7.37 (d, \(J = 9.2\) Hz, 1H), 6.87 (dd, \(J = 9.2, 6.5\) Hz, 1H), 6.71 (t, \(J = 6.9\) Hz, 1H); \(^{13}\)C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 152.73, 147.41, 135.28, 133.30, 125.27, 122.70, 123.59, 121.47, 119.39, 115.38, 76.30; HRMS(ESI): calculated for C\(_{10}\)H\(_7\)IN\(_3\)S\(^+\) [M+H]\(^+\): 327.9400, found: 327.9398.

1-iodo-3-(pyridin-2-yl)imidazo[1,5-a]pyridine (3by):

From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and picolininaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (215.7 mg, 84%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). \(^1\)H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 9.77 (d, \(J = 7.3\) Hz, 1H), 8.46 (s, 1H), 8.19 (d, \(J = 8.1\) Hz, 1H), 7.62 (t, \(J = 7.6\) Hz, 1H), 7.25 (d, \(J = 9.1\) Hz, 1H), 7.05 (t, \(J = 6.2\) Hz, 1H), 6.77 (dd, \(J = 9.2, 6.4\) Hz, 1H), 6.60 (t, \(J = 6.9\) Hz, 1H); \(^{13}\)C
NMR (101 MHz, CDCl₃) δ: 150.16, 148.08, 137.69, 136.50, 134.52, 126.49, 121.92, 121.85, 121.46, 118.15, 114.28, 75.02.

1-iodo-3-(pyridin-3-yl)imidazo[1,5-a]pyridine (3bz):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and nicotinaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (202.9 mg, 79%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 102-104 °C; IR (KBr) 3037, 1629, 1585, 1494, 1406, 1363, 1263, 1172, 1099, 999, 941, 812, 744, 678 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.94 (s, 1H), 8.56 (d, J = 4.9 Hz, 1H), 8.10 (d, J = 7.2 Hz, 1H), 7.99 (dt, J = 7.9, 2.0 Hz, 1H), 7.35 (dd, J = 8.0, 4.9 Hz, 1H), 7.27 (d, J = 9.2 Hz, 1H), 6.75 (dd, J = 9.2, 6.4 Hz, 1H), 6.58 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 149.78, 148.30, 137.29, 135.30, 133.91, 125.77, 123.84, 121.41, 120.78, 119.12, 114.71, 74.99; HRMS(ESI): calculated for C₁₂H₉I₃N⁺ [M+H]⁺: 321.9836, found: 321.9837.

1-iodo-3-(pyridin-4-yl)imidazo[1,5-a]pyridine (3bba):
From 2-pyridinemethanamine (0.8 mmol, 1 eq.) and isonicotinaldehyde (1.6 mmol, 2 eq.), following the general procedure B, the title compound (210.6 mg, 82%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 15:1 – 3:1). m.p.: 161-163 °C; IR (KBr) 3037, 1626, 1591, 1494, 1409, 1357, 1245, 1141, 985, 947, 833, 742, 676 cm⁻¹; ¹H NMR (400 MHz, CDCl₃) δ: 8.66 (s, 2H), 8.26 (d, J = 7.2 Hz, 1H), 7.64 (d, J = 4.4 Hz, 2H), 7.35 (d, J = 9.2 Hz, 1H), 6.83 (dd, J = 9.2, 6.4 Hz, 1H), 6.68 (t, J = 6.8 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 150.57, 137.49, 136.62, 134.60, 121.83, 121.23, 121.18, 119.39, 115.14, 75.95; HRMS(ESI): calculated for C₁₂H₉I₃N⁺ [M+H]⁺: 321.9836, found: 321.9834.

2-phenyl-1H-benzo[d]imidazole (3c)[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and benzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (174.6 mg, 90%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, DMSO-d₆) δ: 8.28 (d, J = 7.6 Hz, 2H), 7.7-7.62 (m, 2H), 7.61-7.45 (m, 3H), 7.29-7.18 (m, 2H); ¹³C NMR (101 MHz, DMSO-d₆) δ: 151.81, 139.96, 130.70, 130.30, 129.41, 129.22, 126.99, 122.63, 115.60.
2-(o-tolyl)-1H-benzo[d]imidazole (3cb)[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-methylbenzaldehyde (1.75 mmol, 1.75 eq.),
following the general procedure C, the title compound (174.7 mg, 84%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, DMSO-d₆) δ: 7.82-7.73 (m, 1H), 7.63 (s, 2H), 7.39 (s, 3H), 7.22 (s, 2H), 2.62 (s, 3H); ¹³C NMR (101 MHz, DMSO-d₆) δ: 152.41, 137.49, 131.72, 130.53, 129.92, 129.78, 126.42, 122.35, 21.50.

2-(m-tolyl)-1H-benzo[d]imidazole (3cc)[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-methylbenzaldehyde (1.75 mmol, 1.75 eq.),
following the general procedure C, the title compound (178.9 mg, 86%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, DMSO-d₆) δ: 8.04 (s, 1H), 7.99 (d, J = 7.7 Hz, 1H), 7.71-7.54 (m, 2H), 7.45 (t, J = 7.6 Hz, 1H), 7.33 (d, J = 7.6 Hz, 1H), 7.28-7.16 (m, 2H), 2.42 (s, 3H); ¹³C NMR (101 MHz, DMSO-d₆) δ: 151.63, 138.69, 131.18, 130.09, 129.36, 127.57, 124.17, 122.76, 115.46, 21.51.

2-(p-tolyl)-1H-benzo[d]imidazole (3cd)[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-methylbenzaldehyde (1.75 mmol, 1.75 eq.),
following the general procedure C, the title compound (185.1 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, DMSO-d₆) δ: 8.10 (d, J = 7.8 Hz, 2H), 7.61 (dd, J = 6.1, 3.1 Hz, 2H), 7.36 (d, J = 7.9 Hz, 2H), 7.27-7.15 (m, 2H), 2.37 (s, 3H); ¹³C NMR (101 MHz, DMSO-d₆) δ: 151.81, 140.14, 139.64, 129.98, 129.81, 127.72, 126.92, 122.53, 115.39, 21.43.

2-(2-methoxyphenyl)-1H-benzo[d]imidazole (3ce)[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-methoxybenzaldehyde (1.75 mmol, 1.75 eq.),
following the general procedure C, the title compound (185.9 mg, 83%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, DMSO-d₆) δ: 8.34 (dd, J = 7.7, 1.8 Hz, 1H), 7.64 (dd, J = 6.0, 3.2 Hz, 2H), 7.54-7.44 (m, 1H), 7.29-7.17 (m, 3H), 7.13 (t, J = 7.5 Hz, 1H), 4.03 (s, 3H); ¹³C NMR (101 MHz, DMSO-d₆) δ: 157.25, 149.36, 131.81, 130.21, 122.35, 121.35, 118.41, 112.57, 56.23.
2-(4-methoxyphenyl)-1H-benzo[d]imidazole (3cf) \(^{[5]}\):
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-methoxybenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (194.9 mg, 87%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.17 (d, \(J = 8.9\) Hz, 2H), 7.59 (dd, \(J = 6.0, 3.2\) Hz, 2H), 7.19 (dd, \(J = 6.0, 3.2\) Hz, 2H), 7.12 (d, \(J = 8.9\) Hz, 2H), 3.83 (s, 3H); \(^{13}\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 161.13, 151.82, 139.82, 128.54, 123.06, 122.30, 115.18, 114.83, 55.76.

2-(2-fluorophenyl)-1H-benzo[d]imidazole (3cg) \(^{[5]}\):
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-fluorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (186.6 mg, 88%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.31 (t, \(J = 7.8\) Hz, 1H), 7.78 - 7.63 (m, 2H), 7.58 - 7.49 (m, 1H), 7.47 - 7.35 (m, 2H), 7.26 (dd, \(J = 6.1, 3.2\) Hz, 2H); \(^{13}\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 159.98 (d, \(J = 249.9\) Hz), 146.95, 132.24 (d, \(J = 8.6\) Hz), 130.74, 130.71, 125.50 (d, \(J = 3.3\) Hz), 122.80, 118.68, 118.57, 116.94 (d, \(J = 21.7\) Hz); \(^{19}\)F NMR (376 MHz, DMSO-d\(_6\)) \(\delta\): -114.57.

2-(3-fluorophenyl)-1H-benzo[d]imidazole (3ch) \(^{[5]}\):
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-fluorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (192.9 mg, 91%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.05 (d, \(J = 7.8\) Hz, 1H), 8.02 - 7.93 (m, 1H), 7.77 - 7.55 (m, 3H), 7.38 - 7.30 (m, 1H), 7.24 (dd, \(J = 6.1, 3.1\) Hz, 2H); \(^{13}\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 162.93 (d, \(J = 243.6\) Hz), 150.45, 150.42, 133.01, 132.93, 131.60 (d, \(J = 8.4\) Hz), 122.99 (d, \(J = 2.9\) Hz), 117.06 (d, \(J = 21.2\) Hz), 113.61, 113.37; \(^{19}\)F NMR (376 MHz, DMSO-d\(_6\)) \(\delta\): -112.39.

2-(4-fluorophenyl)-1H-benzo[d]imidazole (3ci) \(^{[5]}\):
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-fluorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (197.2 mg, 93%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.34 - 8.16 (m, 2H), 7.68 - 7.56 (m, 2H), 7.46 - 7.34 (m, 2H), 7.27 - 7.15 (m, 2H); \(^{13}\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 163.52 (d, \(J = 247.5\) Hz), 150.90, 129.21 (d, \(J = 8.6\) Hz), 127.29 (d, \(J = 3.0\) Hz), 122.61, 116.40 (d, \(J = 21.9\) Hz); \(^{19}\)F NMR (376 MHz, DMSO-d\(_6\)) \(\delta\): -111.17.
2-(2-chlorophenyl)-1H-benzo[d]imidazole (3c)\textsuperscript{[8]}:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-chlorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (205.2 mg, 90%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 7.98-7.89 (m, 1H), 7.73-7.61 (m, 3H), 7.56-7.49 (m, 2H), 7.29-7.20 (m, 2H); \(^13\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 149.60, 132.56, 132.14, 131.64, 130.82, 130.46, 127.88, 122.70.

2-(3-chlorophenyl)-1H-benzo[d]imidazole (3ck)\textsuperscript{[8]}:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-chlorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (212.0 mg, 93%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.25 (s, 1H), 8.16 (d, \(J = 7.3\) Hz, 1H), 7.69 (s, 1H), 7.63-7.52 (m, 3H), 7.23 (s, 2H); \(^13\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 150.20, 134.25, 132.67, 131.40, 130.00, 126.50, 125.48, 119.54, 111.94.

2-(4-chlorophenyl)-1H-benzo[d]imidazole (3cl)\textsuperscript{[8]}:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-chlorobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (216.6 mg, 95%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.20 (d, \(J = 6.6\) Hz, 2H), 7.69-7.56 (m, 4H), 7.27-7.16 (m, 2H); \(^13\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 150.63, 134.97, 133.01, 132.59, 132.48, 129.53, 129.51, 128.61, 122.78.

2-(2-bromophenyl)-1H-benzo[d]imidazole (3cm)\textsuperscript{[8]}:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-bromobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (247.5 mg, 91%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \(^1\)H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 7.81 (t, \(J = 8.6\) Hz, 2H), 7.73-7.59 (m, 2H), 7.55 (t, \(J = 7.5\) Hz, 1H), 7.45 (t, \(J = 7.7\) Hz, 1H), 7.36-7.11 (m, 2H); \(^13\)C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 150.93, 139.10, 133.88, 132.88, 132.75, 131.83, 128.25, 122.66, 122.07, 120.03, 115.83.
2-(3-bromophenyl)-1H-benzo[d]imidazole (3cn)\[5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-bromobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (250.2 mg, 92%) was obtained as a faint yellow solid. 
$R_f = 0.3$ (petroleum ether / ethyl acetate = 20:1 - 5:1). 
$^1H$ NMR (400 MHz, DMSO-$d_6$) $\delta$: 8.39 (t, $J = 1.9$ Hz, 1H), 8.20 (d, $J = 7.8$ Hz, 1H), 7.69 (d, $J = 8.4$ Hz, 2H), 7.60-7.45 (m, 2H), 7.28-7.18 (m, 2H); $^{13}C$ NMR (101 MHz, DMSO-$d_6$) $\delta$: 150.09, 144.11, 135.48, 132.88, 131.63, 129.36, 125.84, 123.42, 122.74, 119.55, 111.99.

2-(4-bromophenyl)-1H-benzo[d]imidazole (3co)\[5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-bromobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (261.1 mg, 96%) was obtained as a faint yellow solid. 
$R_f = 0.3$ (petroleum ether / ethyl acetate = 20:1 - 5:1). 
$^1H$ NMR (400 MHz, DMSO-$d_6$) $\delta$: 8.13 (d, $J = 7.5$ Hz, 2H), 7.77 (d, $J = 7.5$ Hz, 2H), 7.61 (s, 2H), 7.27-7.16 (m, 2H); $^{13}C$ NMR (101 MHz, DMSO-$d_6$) $\delta$: 150.68, 138.25, 132.98, 132.45, 129.86, 128.83, 123.72, 122.85, 122.76.

2-(2-nitrophenyl)-1H-benzo[d]imidazole (3cp)\[5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 2-nitrobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (207.9 mg, 87%) was obtained as a faint yellow solid. 
$R_f = 0.3$ (petroleum ether / ethyl acetate = 20:1 - 5:1). 
$^1H$ NMR (400 MHz, DMSO-$d_6$) $\delta$: 8.04 (d, $J = 8.1$ Hz, 1H), 7.99 (d, $J = 7.7$ Hz, 1H), 7.87 (t, $J = 7.6$ Hz, 1H), 7.61 (t, $J = 7.8$ Hz, 1H), 7.63 (d, $J = 24.1$ Hz, 2H), 7.26 (s, 2H); $^{13}C$ NMR (101 MHz, DMSO-$d_6$) $\delta$: 149.44, 147.78, 144.09, 137.37, 135.07, 133.10, 131.38, 124.76, 124.71, 123.54, 122.36, 119.65, 112.14.

2-(3-nitrophenyl)-1H-benzo[d]imidazole (3cq)\[5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-nitrobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (212.7 mg, 89%) was obtained as a faint yellow solid. 
$R_f = 0.3$ (petroleum ether / ethyl acetate = 20:1 - 5:1). 
$^1H$ NMR (400 MHz, DMSO-$d_6$) $\delta$: 8.51-8.34 (m, 4H), 7.67 (s, 2H), 7.33-7.15 (m, 2H); $^{13}C$ NMR (101 MHz, DMSO-$d_6$) $\delta$: 155.15, 149.46, 148.80, 148.28, 136.49, 131.29, 130.82, 129.73, 129.09, 127.86, 124.76, 123.46, 121.70.
2-(4-nitrophenyl)-1H-benzo[d]imidazole (3cr)\[^5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-nitrobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (217.5 mg, 91%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \[^1\]H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.98 (s, 1H), 8.57 (d, \(J = 7.8\) Hz, 1H), 8.25 (d, \(J = 8.3\) Hz, 1H), 7.78 (t, \(J = 8.0\) Hz, 1H), 7.63 (s, 2H), 7.25-7.20 (m, 2H); \[^13\]C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 149.49, 148.71, 132.85, 132.15, 130.96, 124.51, 123.33, 121.24, 112.05.

4-(1H-benzo[d]imidazol-2-yl)benzonitrile (3cs)\[^5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-formylbenzonitrile (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (210.2 mg, 96%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \[^1\]H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.34 (d, \(J = 7.6\) Hz, 2H), 8.01 (d, \(J = 7.3\) Hz, 2H), 7.65 (s, 2H), 7.32-7.20 (m, 2H); \[^13\]C NMR (101 MHz, DMSO-d\(_6\)) \(\delta\): 149.83, 136.47, 134.71, 133.40, 133.24, 133.19, 130.27, 127.56, 127.43, 123.27, 119.08, 112.35.

2-(4-(trifluoromethyl)phenyl)-1H-benzo[d]imidazole (3ct)\[^5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-(trifluoromethyl)benzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (256.7 mg, 98%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \[^1\]H NMR (400 MHz, DMSO-d\(_6\)) \(\delta\): 8.41 (d, \(J = 8.1\) Hz, 2H), 7.93 (d, \(J = 8.2\) Hz, 2H), 7.65 (s, 2H), 7.29-7.20 (m, 2H); \(^{19}\)F NMR (376 MHz, DMSO-d\(_6\)) \(\delta\): -61.20.

1-methyl-2-phenyl-1H-benzo[d]imidazole (3cu)\[^5\]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-(trifluoromethyl)benzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (187.2 mg, 90%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). \[^1\]H NMR (400 MHz, CDCl\(_3\)) \(\delta\): 7.98-7.79 (m, 1H), 7.79-7.64 (m, 2H), 7.54-7.41 (m, 3H), 7.37-7.24 (m, 3H), 3.75 (s, 3H); \[^13\]C NMR (101 MHz, CDCl\(_3\)) \(\delta\): 153.71, 142.94, 136.58, 130.18, 129.71, 129.41, 128.67, 122.76, 122.41, 119.75, 109.71, 31.63.
**2-(4-methoxyphenyl)-benzothiazole (3cv)**:[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-methoxybenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (163.9 mg, 68%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.06-7.82 (m, 3H), 7.75 (d, J = 8.0 Hz, 1H), 7.36 (t, J = 7.7 Hz, 1H), 7.23 (t, J = 7.6 Hz, 1H), 6.88 (d, J = 8.5 Hz, 2H), 3.74 (s, 3H); ¹³C NMR (101 MHz, CDCl₃) δ: 167.92, 161.93, 154.22, 134.86, 129.13, 126.41, 126.24, 124.82, 122.83, 121.54, 114.38, 55.46.

![2-(4-methoxyphenyl)-benzothiazole (3cv)]

**2-(3-bromophenyl)-benzothiazole (3cw)**:[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 3-bromobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (257.2 mg, 89%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.19 (s, 1H), 8.11 (d, J = 8.0 Hz, 1H), 8.05 (d, J = 8.1 Hz, 1H), 8.00 (d, J = 7.8 Hz, 1H), 7.73 (d, J = 8.1 Hz, 1H), 7.54 (t, J = 7.7 Hz, 1H), 7.47 (q, J = 7.9 Hz, 2H); ¹³C NMR (101 MHz, CDCl₃) δ: 165.85, 153.79, 135.79, 135.34, 135.08, 134.35, 131.92, 129.61, 127.23, 126.79, 126.29, 123.56, 123.00, 122.84.

![2-(3-bromophenyl)-benzothiazole (3cw)]

**2-(4-bromophenyl)-benzothiazole (3cx)**:[5]:
From benzene-1,2-diamine (1 mmol, 1 eq.) and 4-bromobenzaldehyde (1.75 mmol, 1.75 eq.), following the general procedure C, the title compound (262.9 mg, 91%) was obtained as a faint yellow solid. Rf = 0.3 (petroleum ether / ethyl acetate = 20:1 - 5:1). ¹H NMR (400 MHz, CDCl₃) δ: 8.09 (d, J = 8.1 Hz, 1H), 7.96 (d, J = 8.1 Hz, 2H), 7.91 (d, J = 8.1 Hz, 1H), 7.63 (d, J = 7.5 Hz, 2H), 7.56-7.47 (m, 1H), 7.42 (t, J = 7.6 Hz, 1H); ¹³C NMR (101 MHz, CDCl₃) δ: 166.70, 154.07, 135.04, 132.53, 132.24, 128.91, 126.53, 125.47, 125.44, 123.33, 121.69.
6. References


(5) (a) Liu, Q.; Chen, H.; Li, S.; Guo, Y.; Cao, S.; Zhao, Y. H2O2-promoted inter- and intramolecular C-N bond formation: synthesis of quinazoline derivatives. *ChemistrySelect* **2022**, *7* (22), e202201231. (b) Yamaguchi, E.; Shibahara, F.; Murai, T. 1-Alkynyl- and 1-alkenyl-3-
7. Copies of NMR Spectra for Compounds

$^1$H NMR Spectrum of $3a$

$^{13}$C NMR Spectrum of $3a$
$\text{H NMR Spectrum of 3ab}$

$\text{13C NMR Spectrum of 3ab}$
C NMR Spectrum of 3ac

19F NMR Spectrum of 3ac
$^1$H NMR Spectrum of 3ad

$^{13}$C NMR Spectrum of 3ad
$^{19}$F NMR Spectrum of 3ad

$^1$H NMR Spectrum of 3ae
$^{13}$C NMR Spectrum of 3ae

$^1$H NMR Spectrum of 3af
$^{13}$C NMR Spectrum of 3af

$^1$H NMR Spectrum of 3ag
C NMR Spectrum of 3ag

\[ \text{13C NMR Spectrum of 3ag} \]

H NMR Spectrum of 3ah

\[ \text{1H NMR Spectrum of 3ah} \]
$^{13}$C NMR Spectrum of 3ah

$^1$H NMR Spectrum of 3ai
$^1$H NMR Spectrum of 3aj

$^{13}$C NMR Spectrum of 3ai
$^1$H NMR Spectrum of $3ak$

$^{13}$C NMR Spectrum of $3aj$
C NMR Spectrum of 3ak

$\text{NMR Spectrum of 3al}$
$^1$H NMR Spectrum of 3am
\( ^{13}C \) NMR Spectrum of 3am

\( ^1H \) NMR Spectrum of 3an
$^{13}$C NMR Spectrum of 3an

$^1$H NMR Spectrum of 3ao
C NMR Spectrum of 3ao

15C NMR Spectrum of 3ao

19F NMR Spectrum of 3ao
$^1$H NMR Spectrum of 3ap

$^{13}$C NMR Spectrum of 3ap
$^1$H NMR Spectrum of 3aq

$^{13}$C NMR Spectrum of 3aq
$^1$H NMR Spectrum of 3ar

$^{13}$C NMR Spectrum of 3ar
$^1$H NMR Spectrum of 3as

$^{13}$C NMR Spectrum of 3as
\[ \text{H NMR Spectrum of 3at} \]

\[ \text{C NMR Spectrum of 3at} \]
$^1$H NMR Spectrum of 3au

$^{13}$C NMR Spectrum of 3au
H NMR Spectrum of 3av

13C NMR Spectrum of 3av
$^1$H NMR Spectrum of 3aw

$^{13}$C NMR Spectrum of 3aw
$^1$H NMR Spectrum of 3ax

$^{12}$C NMR Spectrum of 3ax
$^1$H NMR Spectrum of 3ay

$^{13}$C NMR Spectrum of 3ay
$^1$H NMR Spectrum of 3az

$^{13}$C NMR Spectrum of 3az
$^1$H NMR Spectrum of 3aab

$^{13}$C NMR Spectrum of 3aab
H NMR Spectrum of \textit{3aac}

\textbf{\textsuperscript{13}C NMR Spectrum of 3aac}
H NMR Spectrum of 3b

13C NMR Spectrum of 3b
H NMR Spectrum of 3bb

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$^{19}$F NMR Spectrum of 3bb

$^1$H NMR Spectrum of 3bc
$^{13}$C NMR Spectrum of 3bc

$^{19}$F NMR Spectrum of 3bc
H NMR Spectrum of 3bd

\[ \text{H NMR Spectrum of 3bd} \]

\[ \text{13C NMR Spectrum of 3bd} \]
$^{19}$F NMR Spectrum of 3bd

$^1$H NMR Spectrum of 3be

76
$^{13}$C NMR Spectrum of 3be

$^1$H NMR Spectrum of 3bf
$^{13}$C NMR Spectrum of 3bf

$^1$H NMR Spectrum of 3bg
$^{13}$C NMR Spectrum of 3bg

$^1$H NMR Spectrum of 3bh
\( ^{13}\text{C} \) NMR Spectrum of 3bh

\( ^{1}\text{H} \) NMR Spectrum of 3bi
$^{13}$C NMR Spectrum of 3bi

$^1$H NMR Spectrum of 3bj
$^{13}$C NMR Spectrum of 3bj

$^1$H NMR Spectrum of 3bk
$^{13}$C NMR Spectrum of 3bk

$^1$H NMR Spectrum of 3bl
$^{13}$C NMR Spectrum of 3bl

$^1$H NMR Spectrum of 3bm
$^{13}$C NMR Spectrum of 3bm

$^1$H NMR Spectrum of 3bn
C NMR Spectrum of 3bn

1H NMR Spectrum of 3bo
$^{13}$C NMR Spectrum of 3bo

$^{19}$F NMR Spectrum of 3bo
$^1$H NMR Spectrum of 3bp

$^{13}$C NMR Spectrum of 3bp
$^1$H NMR Spectrum of 3br

$^{13}$C NMR Spectrum of 3br
H NMR Spectrum of 3bs

\[ \text{H NMR Spectrum of 3bs} \]

\[ \text{\( ^1\)H NMR Spectrum of 3bs} \]

\[ \text{\( ^1\)C NMR Spectrum of 3bs} \]
$^1$H NMR Spectrum of 3bt

$^{13}$C NMR Spectrum of 3bt
$^1$H NMR Spectrum of $^3$bu

$^{13}$C NMR Spectrum of $^3$bu
$^1$H NMR Spectrum of 3bv

$^{13}$C NMR Spectrum of 3bv
"H NMR Spectrum of 3bw

13C NMR Spectrum of 3bw
H NMR Spectrum of 3bx

\textsuperscript{13}C NMR Spectrum of 3bx

96
$^{1}H$ NMR Spectrum of 3by

$^{13}C$ NMR Spectrum of 3by
$^1$H NMR Spectrum of 3bba

$^{13}$C NMR Spectrum of 3bba
$^1$H NMR Spectrum of 3c

$^{13}$C NMR Spectrum of 3c
$^1$H NMR Spectrum of 3cb

$^{13}$C NMR Spectrum of 3cb
$^{1}H$ NMR Spectrum of 3cc

$^{13}C$ NMR Spectrum of 3cc
$^1$H NMR Spectrum of 3cd

$^{13}$C NMR Spectrum of 3cd
$^{1}H$ NMR Spectrum of 3ce

$^{13}C$ NMR Spectrum of 3ce
H NMR Spectrum of 3cf

1H NMR Spectrum of 3cf

13C NMR Spectrum of 3cf
$^1$H NMR Spectrum of 3cg

$^{13}$C NMR Spectrum of 3cg
$^{19}F$ NMR Spectrum of 3cg

$^1H$ NMR Spectrum of 3ch
C NMR Spectrum of 3ch

$^{13}$C NMR Spectrum of 3ch

F NMR Spectrum of 3ch

$^{19}$F NMR Spectrum of 3ch
H NMR Spectrum of 3ci

^1^H NMR Spectrum of 3ci

13C NMR Spectrum of 3ci
$^{19}$F NMR Spectrum of 3ci

$^1$H NMR Spectrum of 3cj
$^{13}\text{C}$ NMR Spectrum of 3cj

$^1\text{H}$ NMR Spectrum of 3ck
$^{13}$C NMR Spectrum of 3ck

$^1$H NMR Spectrum of 3cl
$^{13}$C NMR Spectrum of 3cI

$^1$H NMR Spectrum of 3cm
$^{13}\text{C}$ NMR Spectrum of 3cm

$^1\text{H}$ NMR Spectrum of 3cn
$^1$H NMR Spectrum of 3co

$^{13}$C NMR Spectrum of 3cn
$^{13}$C NMR Spectrum of 3co

$^1$H NMR Spectrum of 3cp
$^{13}$C NMR Spectrum of 3cp

$^1$H NMR Spectrum of 3cq
$^{13}$C NMR Spectrum of 3cq

$^1$H NMR Spectrum of 3cr
$^{13}$C NMR Spectrum of $3cr$

$^1$H NMR Spectrum of $3cs$
$^{13}$C NMR Spectrum of 3cs

$^1$H NMR Spectrum of 3ct
$^{13}$C NMR Spectrum of 3ct

$^{19}$F NMR Spectrum of 3ct
$^{1}$H NMR Spectrum of 3cu

$^{13}$C NMR Spectrum of 3cu
$^1$H NMR Spectrum of 3cv

$^{13}$C NMR Spectrum of 3cv
H NMR Spectrum of 3cw

$^1$H NMR Spectrum of 3cw

$^{13}$C NMR Spectrum of 3cw
$^1$H NMR Spectrum of 3cx

$^{13}$C NMR Spectrum of 3cx