

Electronic Supporting Information for

**I₂-Mediated Coupling of Quinazolinone Enamines with 2-Aminopyridines: A
New Strategy to Access Spiroquinazolinones**

Jingxi Fang, Zhentao Pan,* Tong Liu, Yingbo Rao, Huajiang, Jiang and Yongmin
Ma*

Institute of Advanced Studies and School of Pharmaceutical Sciences, Taizhou
University, Jiaojiang 318000, P R China.

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1. General information.

All starting materials and reagents were purchased from commercial sources and used as received unless otherwise noted. Reactions were monitored using thin-layer chromatography (TLC) on commercial silica gel plates. Visualization of the developed plates was performed under UV light (254 nm). NMR spectra data were obtained on Avance (III) HD 400 MHz instruments. ¹H NMR and ¹³C NMR spectra were referenced to residual protic solvent peaks or TMS signal (0 ppm). ¹⁹F NMR chemical shifts were externally referenced to CCl₃F (0 ppm). Data for ¹H NMR are recorded as follows: chemical shift (δ, ppm), multiplicity (s = singlet, d = doublet, t = triplet, m = multiplet or unresolved, br = broad singlet, coupling constant (s) in Hz, integration). Data for ¹³C and ¹⁹F NMR are reported in terms of chemical shift (δ, ppm). HRMS Spectra were obtained with Waters Q-TOF Premier (ESI, positive mode) spectrometers

2. Experimental section

2.1 General synthesis of spiroquinazolinone iodide salts **3** or imidazo[1,2-*a*]pyridines **4**.

A solution of quinazolinone enamines **1** (1.0 mmol), 2-aminopyridines **2** (1.2 mmol) and molecular iodine (1.2 mmol) were dissolved in CHCl₃ (5 mL) under an ambient atmosphere. The mixture was stirred at 25 °C for 2 h. Then the reaction mixture was concentrated in vacuo and purified by flash column chromatography to afford the corresponding product **3** or **4**.

2.2 Gram-scale preparation of spiroquinazolinone iodide salt **3m**.

A solution of 4-methylene-3-phenethyl-3,4-dihydroquinazolin-2(1H)-one **1m** (1.32g, 5 mmol), 2-aminopyridine **2a** (0.56g, 6 mmol) and molecular iodine (1.52g, 6 mmol) were dissolved in CHCl₃ (25 mL) under an ambient atmosphere. The mixture was stirred at 25 °C for 2 h. Then the reaction mixture was concentrated in vacuo and purified by flash column chromatography to afford the 2'-oxo-3'-phenethyl-1,2,2',3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-3,4'-quinazolin]-4-ium iodide **3m** (1.94g, 80%) as white solid.

2.3 Iodine ion detection experiment

A solution of **3p** (100 mg) and starch (10 mg) were dissolved in H₂O (2 mL). Concentrated sulfuric acid (0.5 mL) was added into the mixture and the color of solution was changed from colorless to blue, proving iodide was the anion in the product **3p**.

2.4 Quantitative analysis

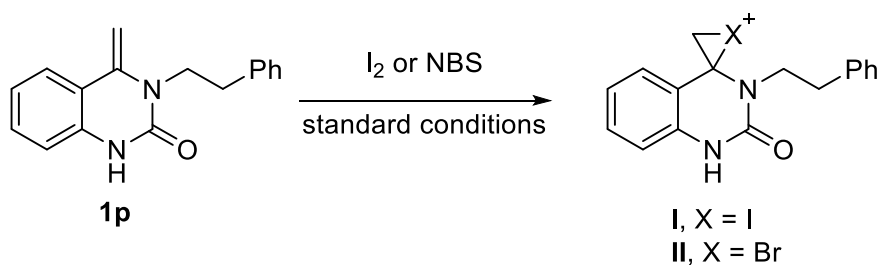
A stock solution of AgNO₃ (0.2 mol L⁻¹) was prepared by dissolving AgNO₃ (509.7 mg, 3 mmol) in 15 mL of deionized water. After product **3p** (484.1 mg, 1 mmol) was dissolved in 5 mL of deionized water, 0.2 mol L⁻¹ AgNO₃ was loaded into a 25 mL burette and added dropwise into the solution until the yellow precipitate was not increase. The consumed volume of titrant was 5.3

mL. Then the reaction mixture was filtered by a Buchner funnel and the filter cake was subsequently washed with water (3 x 0.5 mL). The yellow solid AgI was dried under vacuo and measured to calculate the contents of iodides in the sample using the following equation:

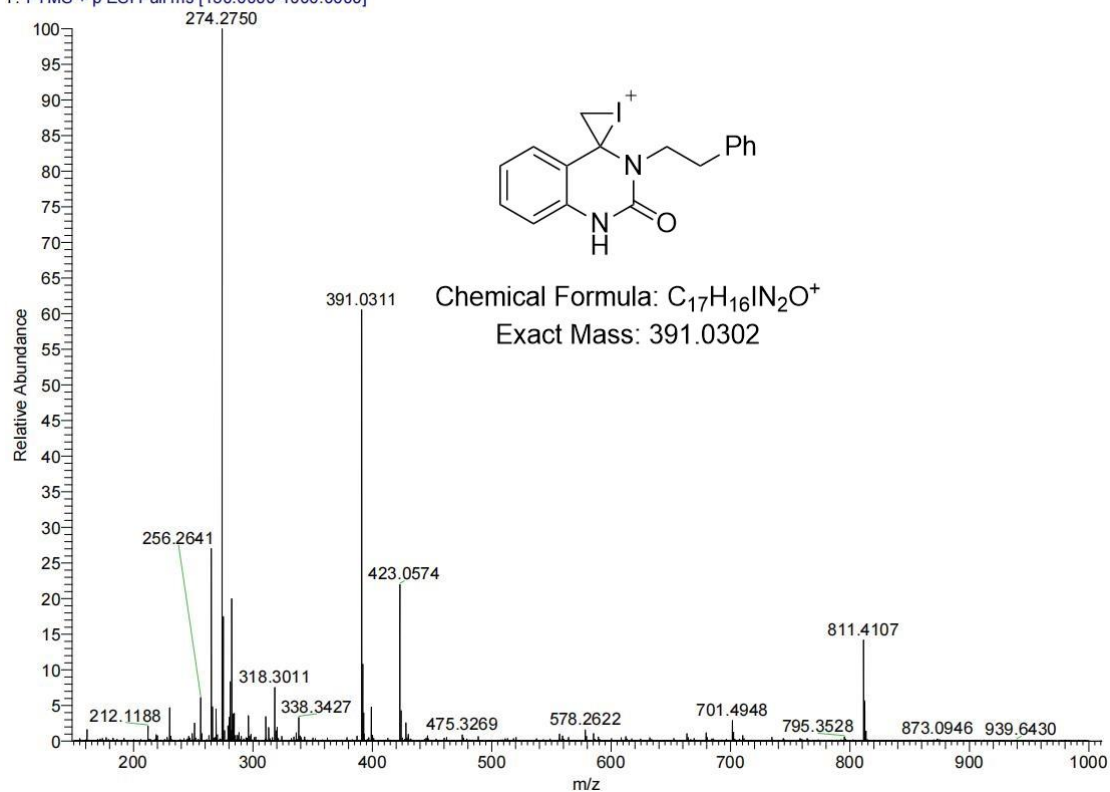
$$W = \frac{m_{\text{AgI}}}{233.8 \times n} \times 100\% = \frac{224.5}{233.8 \times 1} \times 100\% = 96\%$$

where m_{AgI} is the weight of silver iodide obtained, n is the molar amount of **3p** added, and 233.8 is the molecular weight of silver iodide.

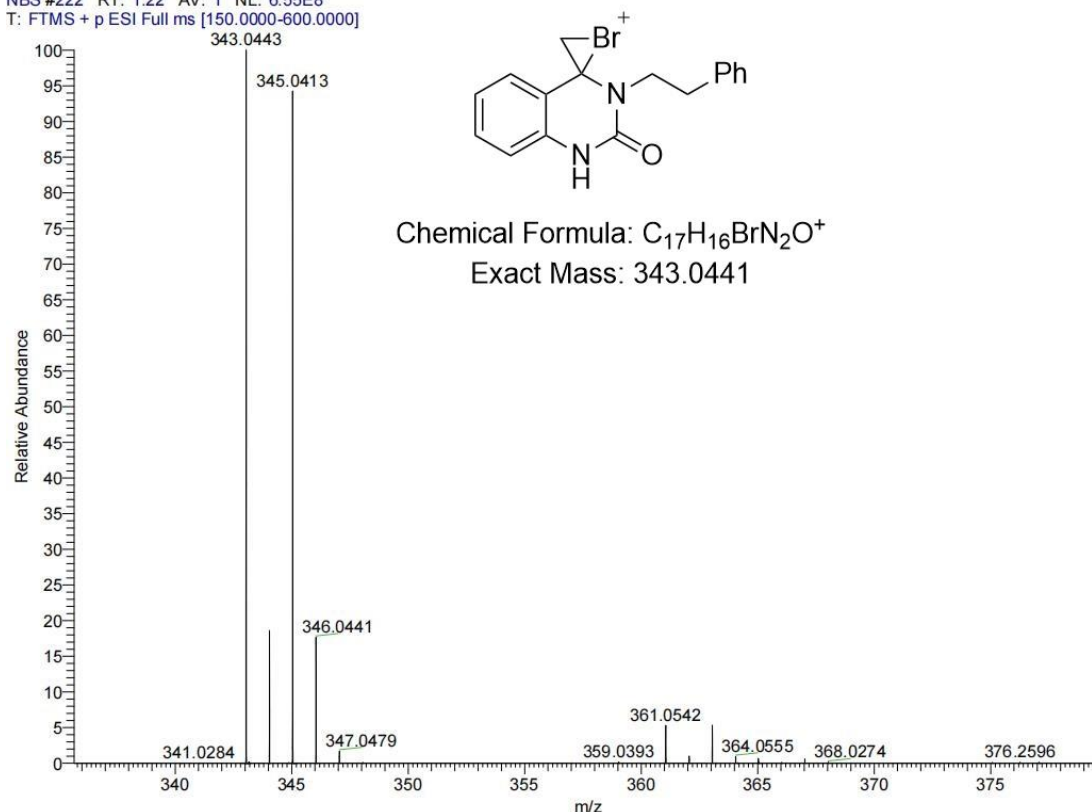
3. HRMS Analysis



FJX3 #70-282 RT: 0.83-2.95 AV: 106 NL: 1.94E8
T: FTMS + p ESI Full ms [150.0000-1000.0000]

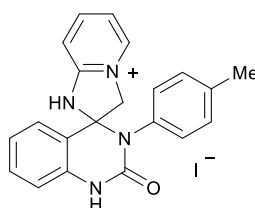


NBS #222 RT: 1.22 AV: 1 NL: 6.55E8
T: FTMS + p ESI Full ms [150.0000-600.0000]



4. Characterization data of products

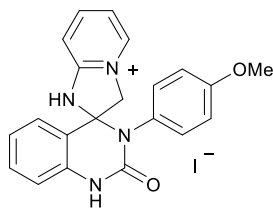
2'-Oxo-3'-(*p*-tolyl)-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (**3a**)



3a

The product **3a** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.55) as a white solid (370 mg, 0.78 mmol, 78%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.75 (s, 1H), 10.29 (s, 1H), 7.98 (d, *J* = 6.4 Hz, 1H), 7.91 – 7.84 (m, 1H), 7.71 (d, *J* = 8.0 Hz, 1H), 7.45 – 7.35 (m, 2H), 7.15 – 7.06 (m, 3H), 7.00 (dd, *J* = 11.2, 8.0 Hz, 2H), 6.90 – 6.83 (m, 2H), 5.35 (d, *J* = 14.8 Hz, 1H), 4.96 (d, *J* = 14.8 Hz, 1H), 2.19 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 152.7, 150.0, 145.7, 138.2, 136.8, 136.7, 133.9, 131.3, 131.2, 131.1, 129.7, 129.6, 128.5, 122.3, 118.1, 114.7, 114.6, 108.4, 80.5, 63.7, 21.0. HRMS-ESI: calcd for C₂₁H₁₉N₄O⁺ [M]⁺: 343.1553, found: 343.1557.

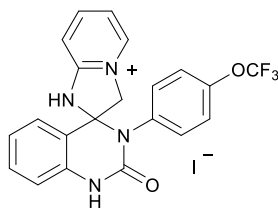
3'-(4-Methoxyphenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (**3b**)



3b

The product **3b** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.45) as a white solid (355 mg, 0.73 mmol, 73%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.28 (s, 1H), 7.96 (d, *J* = 6.4 Hz, 1H), 7.85 (t, *J* = 8.0 Hz, 1H), 7.69 (d, *J* = 8.0 Hz, 1H), 7.41 (q, *J* = 8.0, 6.4 Hz, 2H), 7.11 (dt, *J* = 15.2, 8.0 Hz, 2H), 6.97 (d, *J* = 8.0 Hz, 1H), 6.87 – 6.75 (m, 4H), 5.31 (d, *J* = 14.8 Hz, 1H), 4.92 (d, *J* = 14.8 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 159.2, 152.9, 150.2, 145.5, 136.7, 136.6, 132.6, 132.4, 131.1, 129.0, 128.4, 122.3, 118.4, 114.5, 114.4, 108.5, 80.9, 63.6, 55.8. HRMS-ESI: calcd for C₂₁H₁₉N₄O₂⁺ [M]⁺: 359.1503, found: 359.1502.

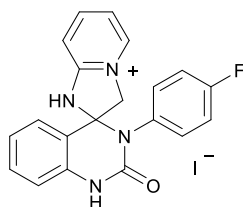
2'-Oxo-3'-(4-(trifluoromethoxy)phenyl)-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3c)



3c

The product **3c** was obtained by flash column chromatography (SiO₂, EA/MeOH = 10/1, R_f = 0.65) as a white solid (400 mg, 0.74 mmol, 74%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.70 (s, 1H), 10.41 (s, 1H), 8.00 (d, *J* = 6.4 Hz, 1H), 7.89 (t, *J* = 8.0 Hz, 1H), 7.74 (dd, *J* = 16.0, 8.4 Hz, 2H), 7.44 (t, *J* = 7.6 Hz, 1H), 7.37 (d, *J* = 8.8 Hz, 1H), 7.27 (dd, *J* = 14.4, 8.8 Hz, 2H), 7.12 (t, *J* = 7.6 Hz, 1H), 7.01 (d, *J* = 8.0 Hz, 1H), 6.88 (dd, *J* = 12.0, 7.6 Hz, 2H), 5.43 (d, *J* = 14.8 Hz, 1H), 4.96 (d, *J* = 14.8 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 152.7, 149.9, 148.3, 145.8, 136.7, 136.6, 135.8, 133.6, 133.5, 131.3, 128.5, 122.4, 122.1, 122.0, 120.3 (q, *J* = 255 Hz), 117.8, 114.9, 114.7, 108.5, 80.8, 63.8. ¹⁹F NMR (376 MHz, DMSO-*d*₆) δ -57.01. HRMS-ESI: calcd for C₂₁H₁₆F₃N₄O₂⁺ [M]⁺: 413.1220, found: 413.1217.

3'-(4-Fluorophenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3d)

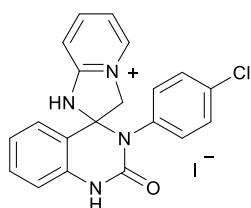


3d

The product **3d** was obtained by flash column chromatography (SiO₂, EA/MeOH = 10/1, R_f = 0.65) as a white solid (422 mg, 0.89 mmol, 89%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.69 (s, 1H), 10.37 (s,

1H), 8.01 (d, $J = 6.3$ Hz, 1H), 7.91 (t, $J = 8.1$ Hz, 1H), 7.74 (d, $J = 7.9$ Hz, 1H), 7.61 (t, $J = 6.8$ Hz, 1H), 7.43 (t, $J = 7.6$ Hz, 1H), 7.27 (t, $J = 6.4$ Hz, 1H), 7.12 (dq, $J = 17.2, 8.8$ Hz, 3H), 7.00 (d, $J = 8.0$ Hz, 1H), 6.90 (d, $J = 8.4$ Hz, 2H), 5.40 (d, $J = 14.8$ Hz, 1H), 4.97 (d, $J = 14.8$ Hz, 1H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 161.9 (d, $J = 245$ Hz), 152.7, 150.0, 145.9, 136.7, 13.7 (d, $J = 12$ Hz), 133.6 (d, $J = 12$ Hz), 132.8 (d, $J = 4$ Hz), 131.5, 128.5, 122.4, 118.0, 116.2 (d, $J = 9$ Hz), 116.0 (d, $J = 9$ Hz), 114.9, 114.7, 108.4, 80.7, 63.7. ^{19}F NMR (376 MHz, DMSO- d_6) δ -108.40. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{FN}_4\text{O}^+$ [M] $^+$: 347.1303, found: 347.1304.

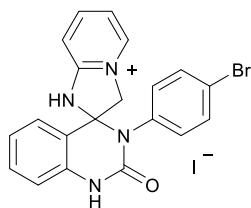
3'-(4-Chlorophenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3e)



3e

The product **3e** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 10/1, $R_f = 0.65$) as a white solid (417 mg, 0.85 mmol, 85%). ^1H NMR (400 MHz, DMSO- d_6) δ 10.70 (s, 1H), 10.31 (s, 1H), 8.01 (d, $J = 6.4$ Hz, 1H), 7.91 (t, $J = 8.0$ Hz, 1H), 7.74 (d, $J = 8.0$ Hz, 1H), 7.51 – 7.39 (m, 4H), 7.17 (d, $J = 8.4$ Hz, 1H), 7.08 (t, $J = 7.6$ Hz, 1H), 6.97 (d, $J = 8.0$ Hz, 1H), 6.89 (t, $J = 7.2$ Hz, 2H), 5.39 (d, $J = 14.8$ Hz, 1H), 4.97 (d, $J = 14.8$ Hz, 1H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 152.6, 149.8, 146.0, 136.6, 136.5, 135.8, 133.7, 133.6, 132.3, 132.2, 131.9, 131.3, 128.5, 122.5, 122.2, 117.9, 114.9, 114.7, 108.4, 80.5, 63.7. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{ClN}_4\text{O}^+$ [M] $^+$: 363.1007, found: 363.1011

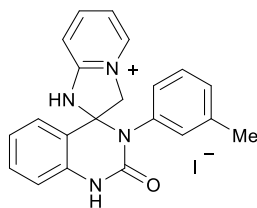
3'-(4-Bromophenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3f)



3f

The product **3f** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 10/1, $R_f = 0.65$) as a white solid (460 mg, 0.86 mmol, 86%). ^1H NMR (400 MHz, DMSO- d_6) δ 10.69 (s, 1H), 10.39 (s, 1H), 8.03 (d, $J = 6.4$ Hz, 1H), 7.92 (dd, $J = 9.2, 7.2$ Hz, 1H), 7.73 (d, $J = 8.0$ Hz, 1H), 7.59 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.43 (t, $J = 7.6$ Hz, 1H), 7.37 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.32 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.24 (dd, $J = 8.0, 2.4$ Hz, 1H), 7.11 (t, $J = 7.6$ Hz, 1H), 6.99 (d, $J = 8.0$ Hz, 1H), 6.91 (d, $J = 8.0$ Hz, 2H), 5.39 (d, $J = 16.0$ Hz, 1H), 4.97 (d, $J = 14.8$ Hz, 1H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 152.7, 149.9, 146.0, 136.8, 136.6, 135.6, 133.5, 133.4, 133.3, 131.3, 129.4, 129.3, 128.4, 122.4, 118.0, 114.9, 114.7, 108.5, 80.6, 63.7. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{BrN}_4\text{O}^+$ [M] $^+$: 407.0502, found: 407.0500.

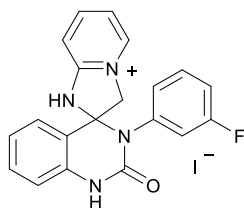
2'-Oxo-3'-(*m*-tolyl)-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3g)



3g

The product **3g** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (376 mg, 0.80 mmol, 80%, *d.r.* = 1:1). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.65 (s, 1H), 10.34 (s, 1H), 8.02 and 7.96 (d, *J* = 6.4 Hz, 1H), 7.91 – 7.84 (m, 1H), 7.73 (t, *J* = 7.2 Hz, 1H), 7.42 (t, *J* = 7.6 Hz, 1H), 7.36 – 7.26 (m, 1H), 7.20 – 7.07 (m, 2H), 7.03 – 6.98 (m, 3H), 6.93 – 6.81 (m, 2H), 5.41 and 5.32 (d, *J* = 14.8 Hz, 1H), 4.97 and 4.94 (d, *J* = 4.4 Hz, 1H), 2.22 and 2.14 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 152.9, 152.7, 149.9, 149.9, 145.7, 138.8, 138.5, 136.9, 136.8, 136.7, 136.6, 136.4, 132.1, 131.7, 131.2, 129.3, 129.2, 129.1, 128.9, 128.5, 128.5, 128.3, 122.3, 118.0, 117.8, 114.8, 114.6, 108.4, 108.3, 80.7, 80.6, 63.8, 63.6, 21.2, 21.1. HRMS-ESI: calcd for C₂₁H₁₉N₄O⁺ [M]⁺: 343.1553, found: 343.1557.

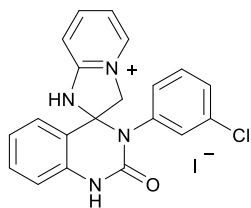
3'-(3-Fluorophenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3h)



3h

The product **3h** was obtained by flash column chromatography (SiO₂, EA/MeOH = 10/1, R_f = 0.60) as a white solid (389 mg, 0.82 mmol, 82%, *d.r.* = 1:1). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.72 and 10.64 (s, 1H), 10.38 (s, 1H), 8.05 (d, *J* = 5.6 Hz, 1H), 7.90 (d, *J* = 9.2 Hz, 1H), 7.75 (d, *J* = 8.0 Hz, 1H), 7.55 – 7.38 (m, 2H), 7.36 – 7.23 (m, 1H), 7.11 (t, *J* = 8.0 Hz, 3H), 7.00 (d, *J* = 8.0 Hz, 1H), 6.92 (dt, *J* = 20.4, 8.0 Hz, 2H), 5.45 (d, *J* = 15.2 Hz, 1H), 4.97 (d, *J* = 15.2 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 162.2 (d, *J* = 245 Hz), 162.1 (d, *J* = 245 Hz), 152.8, 149.8, 145.9, 138.3 (d, *J* = 10 Hz), 136.7, 136.6, 131.3, 130.7 (d, *J* = 8 Hz), 130.6 (d, *J* = 8 Hz), 128.5, 127.9 (d, *J* = 20 Hz), 122.5, 118.8 (d, *J* = 22 Hz), 118.0 (d, *J* = 5 Hz), 116.0, 115.8, 115.0 (d, *J* = 7 Hz), 114.7, 108.5 (d, *J* = 5 Hz), 80.7, 63.9, 63.6. ¹⁹F NMR (376 MHz, DMSO-*d*₆) δ -112.06, -112.55. HRMS-ESI: calcd for C₂₀H₁₆FN₄O⁺ [M]⁺: 347.1303, found: 347.1304.

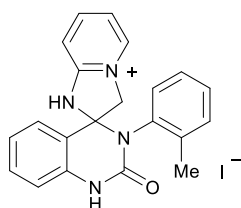
3'-(3-Chlorophenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3i)



3i

The product **3i** was obtained by flash column chromatography (SiO₂, EA/MeOH = 10/1, R_f = 0.60) as a white solid (358 mg, 0.73 mmol, 73%, *d.r.* = 1:1). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.38 (s, 1H), 8.05 (dd, *J* = 12.0, 6.4 Hz, 1H), 7.91 (q, *J* = 9.2 Hz, 1H), 7.78 – 7.53 (m, 2H), 7.42 (t, *J* = 7.6 Hz, 1H), 7.35 – 7.21 (m, 3H), 7.11 (t, *J* = 7.6 Hz, 1H), 6.99 (d, *J* = 8.0 Hz, 1H), 6.90 (dt, *J* = 21.6, 8.0 Hz, 2H), 5.43 (dd, *J* = 22.4, 14.8 Hz, 1H), 4.95 (t, *J* = 15.6 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 153.1, 152.8, 149.9, 149.8, 145.9, 144.1, 144.1, 142.1, 138.1, 138.0, 137.7, 136.9, 136.7, 136.5, 133.5, 133.4, 133.2, 131.6, 131.4, 131.3, 131.0, 130.8, 130.7, 130.5, 130.3, 129.0, 128.6, 128.5, 128.4, 127.3, 126.0, 122.5, 121.9, 121.4, 120.7, 118.6, 118.1, 117.8, 117.6, 117.0, 115.0, 114.7, 113.6, 110.7, 108.6, 108.2, 80.7, 80.6, 63.8. HRMS-ESI: calcd for C₂₀H₁₆ClN₄O⁺ [M]⁺: 363.1007, found: 363.1011.

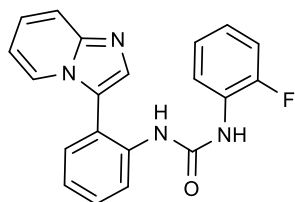
2'-Oxo-3'-(o-tolyl)-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3j**)**



3j

The product **3j** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (254 mg, 0.54 mmol, 54%, *d.r.* > 99:1). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.90 (s, 1H), 10.45 (s, 1H), 7.98 (t, *J* = 8.0 Hz, 2H), 7.74 (d, *J* = 8.0 Hz, 1H), 7.43 (t, *J* = 7.6 Hz, 1H), 7.30 (d, *J* = 7.6 Hz, 1H), 7.20 – 7.09 (s, 3H), 7.06 – 6.95 (m, 3H), 6.87 (t, *J* = 6.8 Hz, 1H), 5.22 (d, *J* = 15.2 Hz, 1H), 4.64 (d, *J* = 15.2 Hz, 1H), 2.20 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 152.2, 149.5, 146.1, 138.0, 136.8, 136.5, 135.6, 131.5, 131.2, 130.7, 129.1, 128.3, 126.9, 122.6, 118.2, 114.8, 114.6, 108.4, 80.4, 61.0, 18.5. HRMS-ESI: calcd for C₂₁H₁₉N₄O⁺ [M]⁺: 343.1553, found: 343.1557.

1-(2-Fluorophenyl)-3-(2-(imidazo[1,2-*a*]pyridin-3-yl)phenyl)urea (4k**)**

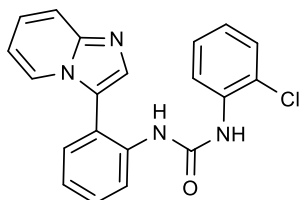


4k

The product **4k** was obtained by flash column chromatography (SiO₂, hexanes/EA = 5/1, R_f = 0.50) as a white solid (235 mg, 0.76 mmol, 76%). ¹H NMR (400 MHz, CDCl₃) δ 11.97 (s, 1H), 8.41 (d, *J* = 9.6

Hz, 1H), 8.17 (t, $J = 7.6$ Hz, 2H), 7.89 (s, 1H), 7.65 (dd, $J = 7.6, 1.6$ Hz, 1H), 7.55 (d, $J = 9.2$ Hz, 1H), 7.35 (ddd, $J = 8.4, 7.2, 1.6$ Hz, 1H), 7.23 (d, $J = 1.2$ Hz, 1H), 7.15 – 6.97 (m, 4H), 6.89 (d, $J = 6.8$ Hz, 2H). ^{13}C NMR (100 MHz, CDCl_3) δ 152.8 (d, $J = 239$ Hz), 152.6, 145.1, 144.0, 137.8, 129.1, 127.3, 125.5, 125.4, 124.6, 123.0 (d, $J = 8$ Hz), 122.3, 121.7, 120.8, 119.6, 117.0, 114.9 (d, $J = 3$ Hz), 114.7 (d, $J = 2$ Hz), 113.3, 109.0. ^{19}F NMR (376 MHz, CDCl_3) δ -131.66. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{FN}_4\text{O}$ [$\text{M} + \text{H}$] $^+$: 347.1303, found: 347.1308.

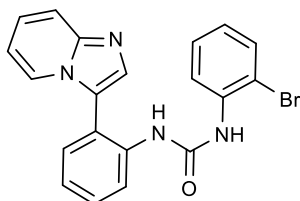
1-(2-Chlorophenyl)-3-(2-(imidazo[1,2-*a*]pyridin-3-yl)phenyl)urea (4l)



4l

The product **4l** was obtained by flash column chromatography (SiO_2 , hexanes/EA = 5/1, $R_f = 0.50$) as a white solid (264 mg, 0.73 mmol, 73%). ^1H NMR (400 MHz, CDCl_3) δ 12.29 (s, 1H), 8.43 (d, $J = 8.4$ Hz, 1H), 8.28 (dd, $J = 8.4, 1.2$ Hz, 1H), 8.16 (d, $J = 6.8$ Hz, 1H), 7.88 (s, 1H), 7.66 (dd, $J = 8.0, 1.6$ Hz, 1H), 7.52 (d, $J = 9.2$ Hz, 1H), 7.37 (ddd, $J = 9.2, 7.6, 1.6$ Hz, 2H), 7.29 (d, $J = 1.6$ Hz, 1H), 7.25 – 7.20 (m, 1H), 7.14 (s, 1H), 7.09 – 7.04 (m, 1H), 6.99 (td, $J = 7.6, 1.6$ Hz, 1H), 6.89 – 6.83 (m, 1H). ^{13}C NMR (100 MHz, CDCl_3) δ 152.5, 145.3, 144.0, 137.9, 135.7, 129.0, 129.0, 127.8, 127.2, 125.5, 125.3, 123.2, 122.5, 122.5, 121.1, 120.6, 119.5, 117.1, 113.2, 108.9. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{ClN}_4\text{O}$ [$\text{M} + \text{H}$] $^+$: 363.1007, found: 363.1008.

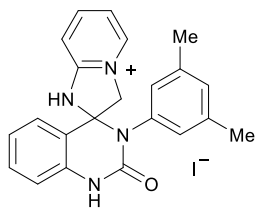
1-(2-Bromophenyl)-3-(2-(imidazo[1,2-*a*]pyridin-3-yl)phenyl)urea (4m)



4m

The product **4m** was obtained by flash column chromatography (SiO_2 , hexanes/EA = 5/1, $R_f = 0.50$) as a white solid (313 mg, 0.77 mmol, 77%). ^1H NMR (400 MHz, CDCl_3) δ 12.41 (s, 1H), 8.44 (dd, $J = 8.4, 1.2$ Hz, 1H), 8.29 (dd, $J = 8.4, 1.6$ Hz, 1H), 8.14 (dt, $J = 6.8, 1.2$ Hz, 1H), 7.87 (s, 1H), 7.65 (dd, $J = 7.6, 1.6$ Hz, 1H), 7.58 – 7.44 (m, 2H), 7.32 (ddt, $J = 8.4, 2.4, 1.2$ Hz, 2H), 7.25 – 7.19 (m, 1H), 7.12 (s, 1H), 7.06 (dd, $J = 7.6, 1.2$ Hz, 1H), 6.96 – 6.89 (m, 1H), 6.85 (dd, $J = 6.8, 1.2$ Hz, 1H). ^{13}C NMR (100 MHz, CDCl_3) δ 152.4, 145.3, 144.0, 137.9, 136.8, 132.2, 129.0, 128.4, 127.1, 125.5, 125.2, 123.7, 122.2, 121.2, 120.5, 119.4, 117.0, 113.2, 113.1, 108.9. HRMS-ESI: calcd for $\text{C}_{20}\text{H}_{16}\text{BrN}_4\text{O}$ [$\text{M} + \text{H}$] $^+$: 407.0502, found: 407.0500.

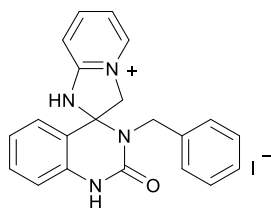
3'-(3,5-Dimethylphenyl)-2'-oxo-1,2',3,3'-tetrahydro-1'*H*-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3n)



3n

The product **3n** was obtained by flash column chromatography (SiO₂, EA/MeOH = 5/1, R_f = 0.45) as a white solid (349 mg, 0.72 mmol, 72%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.58 (s, 1H), 10.30 (s, 1H), 8.01 (d, *J* = 6.4 Hz, 1H), 7.89 (t, *J* = 8.0 Hz, 1H), 7.73 (d, *J* = 8.0 Hz, 1H), 7.41 (t, *J* = 7.6 Hz, 1H), 7.14 – 7.05 (m, 2H), 6.99 (d, *J* = 8.0 Hz, 1H), 6.90 (d, *J* = 8.8 Hz, 1H), 6.83 (t, *J* = 7.2 Hz, 3H), 5.34 (d, *J* = 14.8 Hz, 1H), 4.95 (d, *J* = 14.8 Hz, 1H), 2.18 (s, 3H), 2.09 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 153.0, 149.9, 145.6, 138.5, 138.2, 136.9, 136.7, 136.3, 131.2, 129.9, 129.1, 128.7, 128.5, 122.3, 117.8, 114.8, 114.6, 108.3, 80.7, 63.6, 21.1, 21.0. HRMS-ESI: calcd for C₂₂H₂₁N₄O⁺ [M]⁺: 357.1710, found: 357.1715.

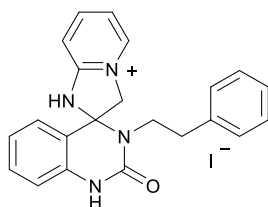
3'-Benzyl-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3o)



3o

The product **3o** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (376 mg, 0.80 mmol, 80%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.22 (s, 1H), 8.14 (d, *J* = 6.4 Hz, 1H), 8.04 (t, *J* = 8.0 Hz, 1H), 7.44 – 7.31 (m, 2H), 7.28 – 7.16 (m, 3H), 7.14 – 7.01 (m, 4H), 6.96 (d, *J* = 8.0 Hz, 1H), 6.87 (d, *J* = 8.8 Hz, 1H), 5.05 (d, *J* = 14.8 Hz, 1H), 4.92 (d, *J* = 14.8 Hz, 1H), 4.75 (d, *J* = 16.8 Hz, 1H), 4.46 (d, *J* = 16.8 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 151.6, 151.1, 146.2, 139.1, 137.5, 135.6, 131.1, 128.8, 127.3, 127.0, 122.7, 119.8, 114.8, 114.4, 108.6, 79.9, 63.5, 45.5. HRMS-ESI: calcd for C₂₁H₁₉N₄O⁺ [M]⁺: 343.1553, found: 343.1557.

2'-Oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3p)

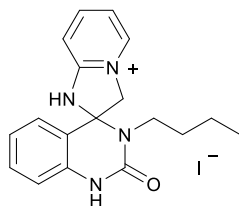


3p

The product **3p** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (402 mg, 0.83 mmol, 83%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.75 (s, 1H), 10.14 (s, 1H),

8.24 (d, $J = 6.4$ Hz, 1H), 8.18 (t, $J = 8.0$ Hz, 1H), 7.48 (d, $J = 8.0$ Hz, 1H), 7.36 (t, $J = 7.6$ Hz, 1H), 7.25 (d, $J = 8.0$ Hz, 3H), 7.21 – 7.13 (m, 2H), 7.05 (dd, $J = 9.6, 7.6$ Hz, 3H), 6.93 (d, $J = 8.0$ Hz, 1H), 5.15 (d, $J = 14.8$ Hz, 1H), 4.95 (d, $J = 14.8$ Hz, 1H), 3.68 – 3.62 (m, 1H), 3.04 – 2.90 (m, 2H), 2.80 – 2.74 (m, 1H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 152.6, 150.1, 146.5, 139.4, 137.8, 135.6, 131.0, 129.0, 128.8, 127.7, 126.9, 122.5, 119.5, 114.8, 114.3, 108.4, 79.9, 64.5, 45.3, 36.1. HRMS-ESI: calcd for $\text{C}_{22}\text{H}_{21}\text{N}_4\text{O}^+$ $[\text{M}]^+$: 357.1710, found: 357.1713.

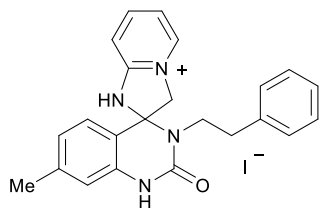
3'-Butyl-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3q)



3q

The product **3q** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 6/1, $R_f = 0.50$) as a white solid (327 mg, 0.75 mmol, 75%). ^1H NMR (400 MHz, DMSO- d_6) δ 10.01 (s, 1H), 8.25 (t, $J = 5.2$ Hz, 1H), 8.16 – 8.04 (m, 1H), 7.41 (d, $J = 8.0$ Hz, 1H), 7.33 (t, $J = 7.6$ Hz, 1H), 7.15 – 7.05 (m, 2H), 7.01 (t, $J = 7.6$ Hz, 1H), 6.89 (d, $J = 8.0$ Hz, 1H), 5.13 (dd, $J = 14.8, 4.0$ Hz, 1H), 4.88 (dd, $J = 14.8, 4.0$ Hz, 1H), 3.47 – 3.41 (m, 1H), 2.82 – 2.75 (m, 1H), 1.68 – 1.58 (m, 1H), 1.51 – 1.41 (m, 1H), 1.23 – 1.03 (m, 2H), 0.74 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 152.9, 150.3, 145.9, 137.6, 137.5, 135.6, 130.8, 127.4, 122.3, 119.9, 114.1, 108.6, 80.2, 64.2, 42.8, 31.8, 20.3, 14.0. HRMS-ESI: calcd for $\text{C}_{18}\text{H}_{21}\text{N}_4\text{O}^+$ $[\text{M}]^+$: 307.1710, found: 307.1713.

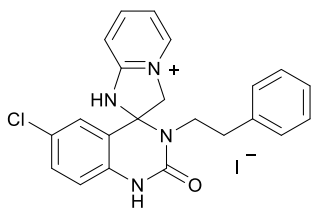
7'-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3r)



3r

The product **3r** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 6/1, $R_f = 0.45$) as a white solid (413 mg, 0.83 mmol, 83%). ^1H NMR (400 MHz, DMSO- d_6) δ 10.07 (s, 1H), 8.20 (d, $J = 6.4$ Hz, 1H), 8.15 (t, $J = 8.0$ Hz, 1H), 7.35 (d, $J = 8.0$ Hz, 1H), 7.28 – 7.17 (m, 4H), 7.11 (t, $J = 6.8$ Hz, 1H), 7.02 (d, $J = 7.2$ Hz, 2H), 6.87 (d, $J = 8.0$ Hz, 1H), 6.71 (s, 1H), 5.09 (d, $J = 14.8$ Hz, 1H), 4.88 (d, $J = 14.8$ Hz, 1H), 3.66 – 3.60 (m, 1H), 3.03 – 2.88 (m, 2H), 2.78 – 2.73 (m, 1H), 2.27 (s, 3H). ^{13}C NMR (100 MHz, DMSO- d_6) δ 152.5, 150.2, 146.5, 140.8, 139.4, 137.7, 135.5, 129.0, 128.8, 127.6, 126.9, 123.4, 116.7, 114.9, 114.2, 108.3, 79.7, 64.4, 45.3, 36.1, 21.3. HRMS-ESI: calcd for $\text{C}_{23}\text{H}_{23}\text{N}_4\text{O}^+$ $[\text{M}]^+$: 371.1866, found: 371.1864.

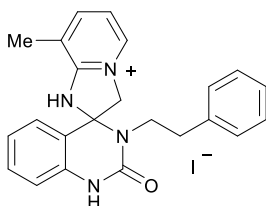
6'-Chloro-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3s)



3s

The product **3s** was obtained by flash column chromatography (SiO₂, EA/MeOH = 8/1, R_f = 0.55) as a white solid (388 mg, 0.75 mmol, 75%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.24 (s, 1H), 8.17 (d, *J* = 6.4 Hz, 1H), 8.11 (t, *J* = 8.0 Hz, 1H), 7.69 (d, *J* = 2.4 Hz, 1H), 7.41 (dd, *J* = 8.4, 2.4 Hz, 1H), 7.28 – 7.21 (m, 3H), 7.18 (t, *J* = 7.2 Hz, 1H), 7.07 (t, *J* = 6.8 Hz, 1H), 7.03 – 6.98 (m, 2H), 6.93 (d, *J* = 8.4 Hz, 1H), 5.09 (d, *J* = 15.2 Hz, 1H), 4.86 (d, *J* = 14.8 Hz, 1H), 3.64 (ddd, *J* = 13.6, 11.6, 5.2 Hz, 2H), 2.98 (td, *J* = 11.6, 4.8 Hz, 1H), 2.86 (td, *J* = 12.8, 11.6, 4.4 Hz, 1H), 2.76 (td, *J* = 11.6, 4.0 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 153.3, 149.9, 145.7, 139.3, 137.5, 134.8, 130.8, 129.0, 128.8, 127.6, 126.8, 125.9, 121.7, 116.0, 113.9, 108.9, 80.1, 64.4, 45.2, 36.2. HRMS-ESI: calcd for C₂₂H₂₀ClN₄O⁺ [M]⁺: 391.1320, found: 391.1315.

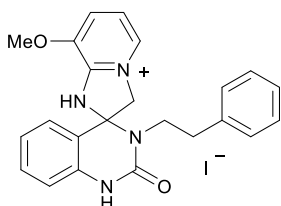
8-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3t)



3t

The product **3t** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (369 mg, 0.74 mmol, 74%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.61 (s, 1H), 10.13 (s, 1H), 8.03 (d, *J* = 31.6 Hz, 2H), 7.48 (d, *J* = 8.0 Hz, 1H), 7.36 (t, *J* = 7.6 Hz, 1H), 7.26 (t, *J* = 7.6 Hz, 2H), 7.18 (t, *J* = 7.2 Hz, 1H), 7.03 (dd, *J* = 22.0, 7.5 Hz, 4H), 6.94 (d, *J* = 8.0 Hz, 1H), 5.14 (d, *J* = 14.8 Hz, 1H), 4.98 (s, 1H), 3.60 (td, *J* = 13.6, 12.4, 4.0 Hz, 1H), 3.07 – 2.90 (m, 2H), 2.76 (dt, *J* = 12.2, 6.4 Hz, 1H), 2.29 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 152.3, 150.1, 139.3, 135.7, 134.7, 131.0, 129.1, 128.7, 127.9, 126.9, 122.5, 119.5, 118.6, 114.6, 114.2, 79.8, 64.7, 45.4, 36.1, 16.4. HRMS-ESI: calcd for C₂₃H₂₃N₄O⁺ [M]⁺: 371.1866, found: 371.1870.

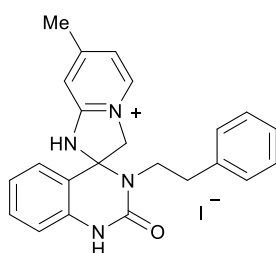
8-Methoxy-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3u)



3u

The product **3u** was obtained by flash column chromatography (SiO₂, EA/MeOH = 5/1, R_f = 0.45) as a white solid (406 mg, 0.79 mmol, 79%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 11.05 (s, 1H), 10.15 (s, 1H), 7.81 (d, *J* = 6.4 Hz, 1H), 7.73 (d, *J* = 8.0 Hz, 1H), 7.49 (d, *J* = 8.0 Hz, 1H), 7.36 (t, *J* = 7.6 Hz, 1H), 7.27 (t, *J* = 7.2 Hz, 2H), 7.19 (t, *J* = 7.2 Hz, 1H), 7.10 (t, *J* = 7.2 Hz, 1H), 7.02 (t, *J* = 8.0 Hz, 3H), 6.92 (d, *J* = 8.0 Hz, 1H), 5.17 (d, *J* = 14.8 Hz, 1H), 5.00 (d, *J* = 14.8 Hz, 1H), 4.03 (s, 3H), 3.64 – 3.58 (m, 1H), 3.03 – 3.00 (m, 1H), 2.96 – 2.86 (m, 1H), 2.78 – 2.73 (m, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 150.0, 146.5, 142.0, 139.5, 135.7, 131.0, 129.1, 128.8, 127.9, 127.80, 126.9, 122.4, 121.7, 119.3, 114.5, 114.2, 80.3, 65.1, 57.7, 45.5, 36.1. HRMS-ESI: calcd for C₂₃H₂₃N₄O₂⁺ [M]⁺: 387.1816, found: 387.1811.

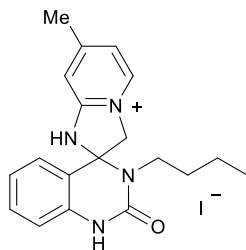
7-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3v)



3v

The product **3j** was obtained by flash column chromatography (SiO₂, EA/MeOH = 5/1, R_f = 0.45) as a white solid (438 mg, 0.88 mmol, 88%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.63 (s, 1H), 10.15 (s, 1H), 8.12 (d, *J* = 6.8 Hz, 1H), 7.44 (d, *J* = 8.0 Hz, 1H), 7.36 (t, *J* = 7.6 Hz, 1H), 7.27 (t, *J* = 7.2 Hz, 2H), 7.19 (t, *J* = 7.2 Hz, 1H), 7.04 (dd, *J* = 13.2, 6.8 Hz, 5H), 6.94 (d, *J* = 8.0 Hz, 1H), 5.08 (d, *J* = 14.8 Hz, 1H), 4.90 (d, *J* = 14.8 Hz, 1H), 3.69 – 3.59 (m, 1H), 3.05 – 2.94 (m, 2H), 2.80 – 2.73 (m, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 159.4, 152.3, 150.2, 139.4, 136.7, 135.6, 131.0, 129.0, 128.9, 127.5, 126.9, 122.5, 119.6, 116.7, 114.3, 107.1, 80.0, 64.0, 45.2, 36.1, 22.2. HRMS-ESI: calcd for C₂₃H₂₃N₄O⁺ [M]⁺: 371.1866, found: 371.1870.

3'-Butyl-7-methyl-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3w)

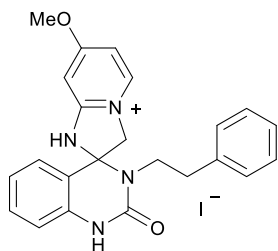


3w

The product **3w** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (374 mg, 0.83 mmol, 83%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.03 (s, 1H), 8.14 (d, *J* = 6.4 Hz, 1H), 7.39 (d, *J* = 8.0 Hz, 1H), 7.33 (t, *J* = 7.6 Hz, 1H), 7.01 (dd, *J* = 16.4, 9.2 Hz, 3H), 6.89 (d, *J* = 8.0 Hz, 1H), 5.09 (d, *J* = 14.6 Hz, 1H), 4.87 (d, *J* = 14.6 Hz, 1H), 3.44 – 3.40 (m, 1H), 2.86 – 2.78

(m, 1H), 2.45 (s, 3H), 1.66 – 1.63 (m, 1H), 1.50 – 1.45 (m, 1H), 1.23 – 1.11 (m, 2H), 0.76 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (100 MHz, $\text{DMSO-}d_6$) δ 159.2, 152.3, 150.3, 136.6, 135.6, 130.9, 127.3, 122.4, 119.7, 116.6, 114.2, 107.0, 80.0, 63.8, 42.8, 31.9, 22.2, 20.3, 14.0. HRMS-ESI: calcd for $\text{C}_{19}\text{H}_{23}\text{N}_4\text{O}^+$ $[\text{M}]^+$: 323.1866 found: 323.1870.

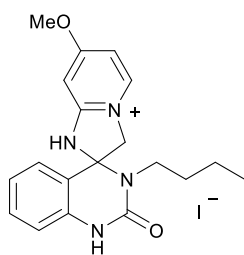
7-Methoxy-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3x)



3x

The product **3x** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 6/1, $R_f = 0.50$) as a white solid (468 mg, 0.91 mmol, 91%). ^1H NMR (400 MHz, $\text{DMSO-}d_6$) δ 10.52 (s, 1H), 10.15 (s, 1H), 8.06 (d, $J = 7.2$ Hz, 1H), 7.42 (d, $J = 8.0$ Hz, 1H), 7.35 (t, $J = 8.0$ Hz, 1H), 7.28 (t, $J = 7.2$ Hz, 2H), 7.19 (t, $J = 7.2$ Hz, 1H), 7.14 – 7.01 (m, 3H), 6.92 (d, $J = 8.0$ Hz, 1H), 6.77 (d, $J = 7.2$ Hz, 1H), 6.57 (s, 1H), 4.96 (d, $J = 14.4$ Hz, 1H), 4.79 (d, $J = 14.4$ Hz, 1H), 4.02 (s, 3H), 3.63 (t, $J = 10.4$ Hz, 1H), 3.00 (t, $J = 10.4$ Hz, 2H), 2.80 (t, $J = 10.0$ Hz, 1H). ^{13}C NMR (100 MHz, $\text{DMSO-}d_6$) δ 171.9, 154.5, 150.2, 139.5, 138.5, 135.6, 131.0, 129.0, 128.9, 127.4, 126.8, 122.4, 119.7, 114.3, 106.4, 88.3, 80.4, 63.5, 58.0, 45.2, 36.1. HRMS-ESI: calcd for $\text{C}_{23}\text{H}_{23}\text{N}_4\text{O}_2^+$ $[\text{M}]^+$: 387.1816, found: 387.1815.

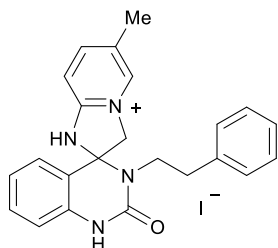
3'-Butyl-7-methoxy-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3y)



3y

The product **3y** was obtained by flash column chromatography (SiO_2 , EA/MeOH = 6/1, $R_f = 0.50$) as a white solid (396 mg, 0.85 mmol, 85%). ^1H NMR (400 MHz, $\text{DMSO-}d_6$) δ 9.98 (s, 1H), 8.11 (d, $J = 7.2$ Hz, 1H), 7.39 – 7.26 (m, 2H), 7.00 (t, $J = 7.6$ Hz, 1H), 6.88 (d, $J = 8.0$ Hz, 1H), 6.74 (dd, $J = 7.2, 2.4$ Hz, 1H), 6.48 (d, $J = 2.4$ Hz, 1H), 4.98 (d, $J = 14.0$ Hz, 1H), 4.76 (d, $J = 14.0$ Hz, 1H), 3.97 (s, 3H), 3.47 – 3.35 (m, 1H), 2.88 – 2.81 (m, 1H), 1.64 – 1.58 (m, 1H), 1.49 – 1.46 (m, 1H), 1.23 – 1.11 (m, 2H), 0.76 (t, $J = 7.2$ Hz, 3H). ^{13}C NMR (100 MHz, $\text{DMSO-}d_6$) δ 171.7, 154.6, 150.4, 138.4, 135.5, 130.9, 127.3, 122.4, 119.9, 114.1, 106.2, 88.3, 80.5, 63.3, 58.0, 42.8, 31.8, 20.3, 14.0. HRMS-ESI: calcd for $\text{C}_{19}\text{H}_{23}\text{N}_4\text{O}_2^+$ $[\text{M}]^+$: 339.1816, found: 339.1815.

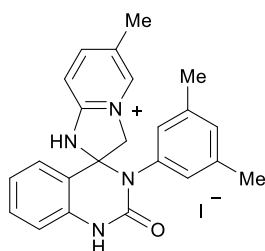
6-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3z)



3z

The product **3z** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (418 mg, 0.84 mmol, 84%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.10 (s, 1H), 8.07 (s, 1H), 8.02 (d, *J* = 9.2 Hz, 1H), 7.41 (d, *J* = 8.0 Hz, 1H), 7.34 (t, *J* = 7.6 Hz, 1H), 7.26 (t, *J* = 7.6 Hz, 2H), 7.20 (t, *J* = 6.8 Hz, 2H), 7.05 (t, *J* = 7.2 Hz, 3H), 6.92 (d, *J* = 8.0 Hz, 1H), 5.06 (d, *J* = 14.8 Hz, 1H), 4.84 (d, *J* = 14.8 Hz, 1H), 3.67 (t, *J* = 10.8 Hz, 1H), 3.01 – 2.89 (m, 2H), 2.79 (t, *J* = 10.8 Hz, 1H), 2.25 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 151.9, 150.4, 147.5, 139.5, 135.5, 135.1, 130.8, 130.7, 129.0, 128.9, 127.4, 126.8, 122.4, 120.4, 114.2, 108.3, 80.5, 64.5, 45.1, 36.2, 17.1. HRMS-ESI: calcd for C₂₃H₂₃N₄O⁺ [M]⁺: 371.1866, found: 371.1870.

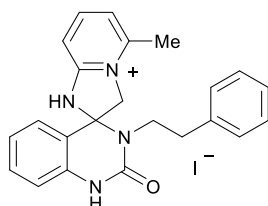
3'-(3,5-Dimethylphenyl)-6-methyl-2'-oxo-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3a')



3a'

The product **3a'** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (408 mg, 0.82 mmol, 82%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.33 (s, 1H), 10.22 (s, 1H), 7.93 (s, 1H), 7.77 (dd, *J* = 9.0, 1.7 Hz, 1H), 7.70 (d, *J* = 7.8 Hz, 1H), 7.39 (t, *J* = 7.7 Hz, 1H), 7.06 (q, *J* = 6.3, 4.4 Hz, 2H), 6.97 (d, *J* = 8.0 Hz, 1H), 6.86 – 6.70 (m, 3H), 5.34 (d, *J* = 14.8 Hz, 1H), 4.90 (d, *J* = 14.8 Hz, 1H), 2.15 (s, 3H), 2.08 (s, 3H), 2.06 (s, 3H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 151.6, 150.0, 147.2, 138.5, 138.2, 136.7, 136.3, 134.3, 131.2, 129.8, 129.1, 128.5, 128.3, 124.2, 122.3, 117.9, 114.6, 108.0, 80.9, 63.9, 21.1, 17.0. HRMS-ESI: calcd for C₂₃H₂₃N₄O⁺ [M]⁺: 371.1866, found: 371.1870.

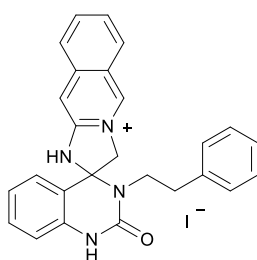
5-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-a]pyridine-2,4'-quinazolin]-4-ium iodide (3b')



3b'

The product **3b'** was obtained by flash column chromatography (SiO₂, EA/MeOH = 8/1, R_f = 0.55) as a white solid (329 mg, 0.66 mmol, 66%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.69 (s, 1H), 10.15 (s, 1H), 8.11 (s, 1H), 7.48 (d, *J* = 8.0 Hz, 1H), 7.37 (s, 1H), 7.27 (t, *J* = 7.2 Hz, 2H), 7.20 (d, *J* = 7.2 Hz, 1H), 7.09 (dd, *J* = 22.0, 8.0 Hz, 5H), 6.94 (d, *J* = 8.0 Hz, 1H), 5.02 (d, *J* = 14.8 Hz, 1H), 4.86 (d, *J* = 14.8 Hz, 1H), 3.80 – 3.79 (m, 1H), 3.17 (s, 3H), 3.03 – 3.00 (m, 1H), 2.91 – 2.85 (m, 1H), 2.79 – 2.74 (m, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 153.1, 150.2, 148.9, 146.5, 139.4, 135.5, 131.0, 129.1, 128.9, 127.7, 126.9, 122.5, 119.8, 115.1, 114.2, 105.1, 79.4, 63.0, 45.3, 36.3, 19.0. HRMS-ESI: calcd for C₂₃H₂₃N₄O⁺ [M]⁺: 371.1866, found: 371.1870.

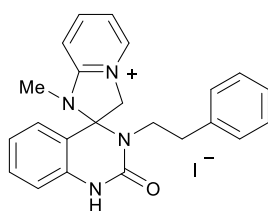
2'-Oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*b*]isoquinoline-2,4'-quinazolin]-4-ium iodide (3c')



3c'

The product **3c'** was obtained by flash column chromatography (SiO₂, EA/MeOH = 8/1, R_f = 0.55) as a yellow solid (395 mg, 0.74 mmol, 74%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.10 (s, 1H), 9.45 (s, 1H), 8.17 (d, *J* = 8.4 Hz, 1H), 8.01 (d, *J* = 8.4 Hz, 1H), 7.89 (s, 1H), 7.53 (t, *J* = 7.6 Hz, 1H), 7.48 (d, *J* = 6.4 Hz, 2H), 7.36 (t, *J* = 7.6 Hz, 1H), 7.14 (dd, *J* = 13.6, 6.8 Hz, 4H), 7.06 – 6.99 (m, 3H), 6.94 (d, *J* = 8.0 Hz, 1H), 5.31 (s, 1H), 5.18 (s, 1H), 3.73 (t, *J* = 11.2 Hz, 1H), 3.09 – 3.01 (m, 2H), 2.83 (t, *J* = 11.2 Hz, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ 150.4, 148.1, 144.0, 142.4, 139.4, 136.3, 135.5, 130.9, 130.2, 128.9, 128.9, 127.4, 126.8, 126.6, 126.1, 122.5, 122.0, 120.2, 114.2, 99.0, 79.5, 65.3, 45.0, 36.2. HRMS-ESI: calcd for C₂₆H₂₃N₄O⁺ [M]⁺: 407.1866, found: 407.1872.

1-Methyl-2'-oxo-3'-phenethyl-1,2',3,3'-tetrahydro-1'H-spiro[imidazo[1,2-*a*]pyridine-2,4'-quinazolin]-4-ium iodide (3d')

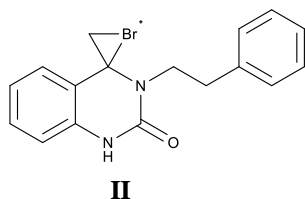


3d'

The product **3d'** was obtained by flash column chromatography (SiO₂, EA/MeOH = 6/1, R_f = 0.50) as a white solid (379 mg, 0.76 mmol, 76%). ¹H NMR (400 MHz, DMSO-*d*₆) δ 10.27 (s, 1H), 8.30 (q, *J* = 8.4, 7.6 Hz, 2H), 7.46 (d, *J* = 8.0 Hz, 1H), 7.38 (t, *J* = 7.6 Hz, 2H), 7.29 – 7.14 (m, 4H), 7.00 (dd, *J* = 26.0, 7.6 Hz, 4H), 5.27 (d, *J* = 15.6 Hz, 1H), 5.09 (d, *J* = 15.2 Hz, 1H), 3.32 – 3.29 (m, 1H), 3.14 – 3.11 (m, 1H), 3.00 – 2.98 (m, 1H), 2.80 (s, 3H), 2.75 – 2.72 (m, 1H). ¹³C NMR (100 MHz, DMSO-*d*₆) δ

152.1, 150.4, 146.7, 139.3, 138.2, 136.8, 131.5, 129.0, 128.9, 127.7, 126.9, 122.8, 116.1, 114.8, 114.5, 107.7, 83.9, 63.1, 45.4, 34.9, 27.5. HRMS-ESI: calcd for $C_{23}H_{23}N_4O^+$ $[M]^+$:371.1866, found: 371.1870.

2'-Oxo-3'-phenethyl-2',3'-dihydro-1'H-spiro[bromirane-2,4'-quinazolin]-1-ium (II)



The product **II** was obtained by flash column chromatography (SiO_2 , hexanes/EA = 5/1, R_f = 0.4) as a white solid (247mg, 0.72 mmol, 72%). 1H NMR (400 MHz, $DMSO-d_6$) δ 9.79 (s, 1H), 7.78 – 7.61 (m, 2H), 7.32 (m, 3H), 7.28 – 7.18 (m, 3H), 7.01 (t, J = 7.6 Hz, 1H), 6.87 (d, J = 8.0 Hz, 1H), 6.45 (s, 1H), 3.80 – 3.56 (m, 2H), 3.01 – 2.73 (m, 2H). ^{13}C NMR (100 MHz, $DMSO-d_6$) δ 152.1, 140.2, 137.4, 130.6, 129.0, 128.9, 128.9, 126.5, 120.6, 118.8, 113.6, 87.6, 54.6, 43.0, 36.3. HRMS-ESI: calcd for $C_{17}H_{16}BrN_2O^+$ $[M]^+$:343.0441, found: 343.0443.

5. 1H NMR, ^{13}C NMR and ^{19}F spectra of products

