Supporting Information for:

**F-element metalated dipyrrins: Synthesis and characterization of a family of uranyl bis(dipyrrinate) complexes**

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+ = protonated ligand
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Figure S34 Solid-state molecular structure of $4^{\text{anis}}$-THF-THF-$\text{C}_5\text{H}_{12}$ with 50% probability ellipsoids. Co-crystallized solvent and second molecule of $4^{\text{anis}}$-THF omitted for clarity. Selected bond lengths (Å) and bond angles (°): U1-O1 = 1.759(4); U1-O2 = 1.764(4); U1-O3 = 2.463(3); U1-N1 = 2.546(5); U1-N2 = 2.453(5); U1-N3 =2.479(5); U1-N4 = 2.503(5); O1-U1-O2 = 176.8(2); N2-N1-C2 = 164.7(4); N1-N2-C1 = 160.0(4); N4-N3-C3 = 161.7(4); C4-N4-N3 = 163.6(4); N4-U1-O1 = 84.2(2); O3-U1-O1 = 83.9(2); N1-U1-O1 = 98.9(2); N2-U1-O1 = 82.0(2); N3-U1-O1 = 97.6(2); N4-U1-O1 = 84.2(2). Dihedral angle of the planes between pyrrole 4 (containing N4) and pyrrole 3 (containing N3) = 29.58°. Dihedral angle of the planes between pyrrole 1 (containing N1) and pyrrole 2(containing N2) = 29.58°.
Figure S35. Solid-state molecular structure of $4^{\text{Fe}}$-THF-C$_5$H$_{12}$ with 50% probability ellipsoids. Co-crystallized solvent molecule omitted for clarity. Selected bond lengths (Å) and bond angles (°): U1-O1 = 1.773(3); U1-O3 = 2.489(5); U1-N1 = 2.457(4); U1-N2 = 2.482(4); O1-U1-O2 = 177.0; N2-N1-C1 = 150.3(4); N1-N2-C2 = 165.5(4); O3-U1-O1 = 89.1; N1-U1-O1 = 91.3; N2-U1-O1 = 89.1; Dihedral angle of the planes between pyrrole 1 (containing N1) and pyrrole 2 (containing N2) = 37.72°.
**Figure S36.** Solid-state molecular structure of 4mes-DMAP·THF·0.5C₅H₁₂ with 50% probability ellipsoids. Co-crystallized solvent omitted for clarity. Selected bond lengths (Å) and bond angles (°): U1-O1 = 1.776(4); U1-O2 = 1.764(4); U1-N5 = 2.489(5); U1-N1 = 2.554(4); U1-N2 = 2.472(5); U1-N3 = 2.556(4); U1-N4 = 2.468(4); O1-U1-O2 = 176.9(2); N2-N1-C1 = 169.2(4); N1-N2-C2 = 164.3(4); N4-N3-C3 = 167.5(4); C4-N4-N3 = 164.8(4); N4-U1-O1 = 100.1(2); N5-U1-O1 = 89.1(2); N1-U1-O1 = 96.4(2); N2-U1-O1 = 82.6(2); N3-U1-O1 = 82.2(2). Dihedral angle of the planes between pyrrole 4 (containing N4) and pyrrole 3 (containing N3) = 22.34°. Dihedral angle of the planes between pyrrole 1 (containing N1) and pyrrole 2 (containing N2) = 20.41°.
Figure S37. Side view of the solid-state molecular structure of $4^{\text{ord}}$-THF-3THF with space filling atoms. Green atom is uranium, red atoms are oxygen, blue atoms are nitrogen, grey atoms are carbon, and hydrogen atoms are white.
Table S1. X-ray crystallographic data for $^4$tolyl-THF·3THF, $^4$anis-THF·THF·C$_5$H$_{12}$, $^4$Fc-THF·C$_5$H$_{12}$, and $^4$mes-DMAP·THF·0.5C$_5$H$_{12}$.

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<tr>
<th>Empirical formula</th>
<th>$^4$tolyl-THF·3THF</th>
<th>$^4$anis-THF·THF·C$<em>5$H$</em>{12}$</th>
<th>$^4$mes-DMAP·THF·0.5C$<em>5$H$</em>{12}$</th>
<th>$^4$Fc-THF·C$<em>5$H$</em>{12}$</th>
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</thead>
<tbody>
<tr>
<td>Crystal Habit, color</td>
<td>Block, red-orange</td>
<td>Plate, red-orange</td>
<td>Plate, red</td>
<td>Plate, dark red</td>
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<tr>
<td>Crystal size (mm)</td>
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<td>0.41 × 0.33 × 0.06</td>
<td>0.96 × 0.63 × 0.19</td>
<td>0.72 × 0.15 × 0.01</td>
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<tr>
<td>Crystal system</td>
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<td>P-1</td>
<td>P2$_1$/c</td>
<td>P-3c1</td>
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<td>4625.7(5)</td>
<td>6141.2(5)</td>
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<tr>
<td>a (Å)</td>
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<td>14.5004(7)</td>
<td>9.3522(6)</td>
<td>20.5164(8)</td>
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<tr>
<td>b (Å)</td>
<td>15.8228(5)</td>
<td>16.3863(8)</td>
<td>26.9204(16)</td>
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<td>c (Å)</td>
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<td>16.8469(7)</td>
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<td>99.0510(10)°</td>
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<td>90°</td>
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<tr>
<td>β(°)</td>
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<td>103.8570(10)°</td>
<td>102.9630(10)°</td>
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<tr>
<td>γ(°)</td>
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<td>98.0330(10)°</td>
<td>90°</td>
<td>120°</td>
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<td>5942.79</td>
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<tr>
<td>Density (calculated) (Mg/m$^3$)</td>
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<td>1.704</td>
<td>1.603</td>
<td>1.607</td>
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<td>Absorption coefficient (mm$^{-1}$)</td>
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<td>F$_{000}$</td>
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<tr>
<td>Unique reflections</td>
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<td>Final R indices [I &gt; 2σ(I)]</td>
<td>R$_1$ = 0.0456, wR$_2$ = 0.1109</td>
<td>R$_1$ = 0.0404, wR$_2$ = 0.1032</td>
<td>R$_1$ = 0.0538, wR$_2$ = 0.1081</td>
<td>R$_1$ = 0.0330, wR$_2$ = 0.0765</td>
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<td>Largest diff. peak and hole (e Å$^{-3}$)</td>
<td>2.36 and -1.19</td>
<td>2.23 and -0.76</td>
<td>2.65 and -5.48</td>
<td>2.97 and -0.82</td>
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<td>GOF</td>
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<td>1.050</td>
<td>1.258</td>
<td>1.142</td>
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Figure S38. Room temperature UV/vis absorption spectra for $2^{\text{tol}}$ (benzene, 24.49 μM) (grey), $3^{\text{tol}}$ (benzene, 10.46 μM) (orange), and $4^{\text{tol}}$-DMAP (benzene, 11.8 μM) (black).
Figure S39. Room temperature UV/vis absorption spectra for $2^{\text{anis}}$ (benzene, 24.58 μM) (grey), $3^{\text{anis}}$ (benzene, 9.68 μM) (orange), and $4^{\text{anis}}$-DMAP (benzene, 11.76 μM) (black).
Figure S40. Room temperature UV/vis absorption spectra for $2^{\text{mes}}$ (benzene, 25.09 μM) (grey), $3^{\text{mes}}$ (benzene, 9.66 μM) (orange), and $4^{\text{mes}}$-DMAP (benzene, 12.9 μM) (black).
Figure S41. Room temperature UV/vis absorption spectra for $2^\text{Fe}$ (benzene, 27.98 μM) (grey), $3^\text{Fe}$ (benzene, 10.67 μM) (orange), and $4^\text{Fe}$-DMAP (benzene, 10.59 μM) (black).
Figure S42. Room temperature UV/vis absorption spectra for protonated dipyrrins $2^\text{tol}$ (purple), $2^\text{anis}$ (red), $2^\text{mes}$ (blue), and $2^\text{Fc}$ (green).
Figure S43. Room temperature UV/vis absorption spectra for sodium dipyrrins $3^{\text{tol}}$ (purple), $3^{\text{anis}}$ (red), $3^{\text{mes}}$ (blue), and $3^{\text{Fe}}$ (green).
Figure S44. Room temperature UV/vis absorption spectra for uranyl dipyrrins $4^{\text{tol}}$-DMAP (purple), $4^{\text{anis}}$-DMAP (red), $4^{\text{mes}}$-DMAP (blue), and $4^{\text{Fe}}$-DMAP (green).
Table S2. UV/vis absorption data.

<table>
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<tr>
<th>Compound</th>
<th>Concentration (μM)</th>
<th>( \lambda_{\text{max}} ) (nm)</th>
<th>( \varepsilon ) (L·mol(^{-1})·cm(^{-1}))</th>
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<td>2(^{\text{tol}})</td>
<td>24.49</td>
<td>436</td>
<td>22832</td>
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<td>2(^{\text{anis}})</td>
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<td>437</td>
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<td>2(^{\text{mes}})</td>
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<td>3(^{\text{tol}})</td>
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<td>4(^{\text{Fe}})-DMAP</td>
<td>10.59</td>
<td>462</td>
<td>27578</td>
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</table>
Figure S45. Room temperature cyclic voltammogram of 4\textsuperscript{th}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte). The scan rate is 250 mV/s.
Figure S46. Room temperature cyclic voltammogram of wave 1 of 4\textsuperscript{tol}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at \(E_{1/2} = 0\) V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte).
Figure S47. Room temperature cyclic voltammogram of wave 2 of 4\textsuperscript{th}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte).
**Figure S48.** Room temperature cyclic voltammogram of wave 3 of 4$^\text{tol}$-DMAP in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2} = 0$ V). (0.1M [NBu$_4$][PF$_6$] as supporting electrolyte).
**Figure S49.** Room temperature cyclic voltammogram of 4\textsuperscript{anil}DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte). The scan rate is 125 mV/s.
Figure S50. Room temperature cyclic voltammogram of wave 1 of 4\textsuperscript{anis}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at $E_{1/2} = 0$ V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte).
Figure S51. Room temperature cyclic voltammogram of wave 2 of 4<sub>anis</sub>-DMAP in THF (vs internally referenced Cp₂Fe/Cp₂Fe⁺ at E<sub>1/2</sub> = 0 V). (0.1M [NBu₄][PF₆] as supporting electrolyte).
Figure S52. Room temperature cyclic voltammogram of wave 3 of 4\textsuperscript{anis}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte).
Figure S53. Room temperature cyclic voltammogram of 4\textsuperscript{mes}DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte). The scan rate is 125 mV/s.
Figure S54. Room temperature cyclic voltammogram of wave 1 of $4^\text{mes}$-DMAP in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2} = 0$ V). (0.1M $[\text{NBu}_4][\text{PF}_6]$ as supporting electrolyte).
Figure S55. Room temperature cyclic voltammogram of wave 2 of 4mes-DMAP in THF (vs internally referenced Cp²Fe/Cp²Fe⁺ at E₁/₂ = 0 V). (0.1M [NBu₄][PF₆] as supporting electrolyte).
Figure S56. Room temperature cyclic voltammogram of wave 3 of 4\textsubscript{mes}-DMAP in THF (vs internally referenced Cp\textsubscript{2}Fe/Cp\textsubscript{2}Fe\textsuperscript{+} at E\textsubscript{1/2} = 0 V). (0.1M [NBu\textsubscript{4}][PF\textsubscript{6}] as supporting electrolyte).
Figure S57. Room temperature cyclic voltammogram of wave 4 of 4^{mes}-DMAP in THF (vs internally referenced Cp₂Fe/Cp₂Fe⁺ at E_{1/2} = 0 V). (0.1M [NBu₄][PF₆] as supporting electrolyte).
Figure S58. Room temperature cyclic voltammogram of $^{4}\text{Fe}$-DMAP in THF at 125 mV (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2} = 0$ V). (0.1M [NBu$_4$][PF$_6$] as supporting electrolyte). The scan rate is 125 mV/s.
Figure S59. Room temperature cyclic voltammogram of 4$^{Fe}$-DMAP in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at E$_{1/2}$=0 V). (0.1M [NBu$_4$][PF$_6$] as supporting electrolyte).
**Figure S60.** Room temperature cyclic voltammogram of $2^\text{anis}$ in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2}=0$ V). (0.1M [NBu$_4$][PF$_6$] as supporting electrolyte).
Figure S61. Room temperature cyclic voltammogram of wave 1 of $2^{\text{anis}}$ in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2}=0$ V). (0.1M [NBu$_4$][PF$_6$] as supporting electrolyte).
Figure S62. Room temperature cyclic voltammograms of UO$_2$(N(Si(CH$_3$)$_3$)$_2$)$_2$(THF)$_2$ in THF (vs internally referenced Cp$_2$Fe/Cp$_2$Fe$^+$ at $E_{1/2} = 0$ V). Bottom voltammogram at a scan rate of 250 mV/s.
Figure S63. Fluorescence spectra of dipyrrins (2), sodium dipyrrins (3), and uranyl bis(dipyrrins) (4) in benzene (10 μM) with excitation wavelengths set to their respective absorption maxima determined in the same solvent and concentrations.
Figure S64. IR spectrum (KBr pellet) of 4-tol-DMAP.
Figure S65. IR spectrum (KBr pellet) of 4-^\text{anis}-\text{DMAP}.
Figure S66. IR spectrum (KBr pellet) of 4$^{\text{mes}}$-DMAP.
Figure S67. IR spectrum (KBr pellet) of $4^\text{Fe}$-DMAP.
Figure S68. IR spectra (KBr pellet) of uranyl bis(dipyrrin) complexes. The arrows indicate the asymmetric $\nu_3$ U-O stretch of uranyl at 963 cm$^{-1}$. 