Supporting Information

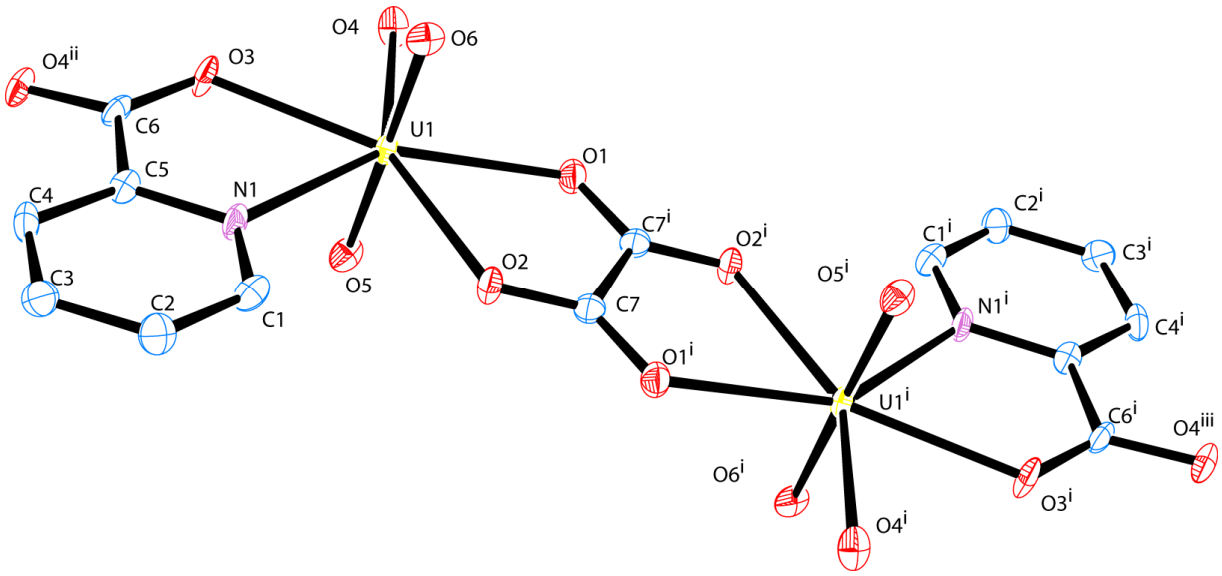


Figure S1. ORTEP illustration of **1**. Ellipsoids shown at 50% probability level. Superscript donates symmetry transformations i =-x+1, -y+1, -z; ii = -x+2, y+1/2, -z+1/2; iii = x-1, -y+1/2, z-1/2.

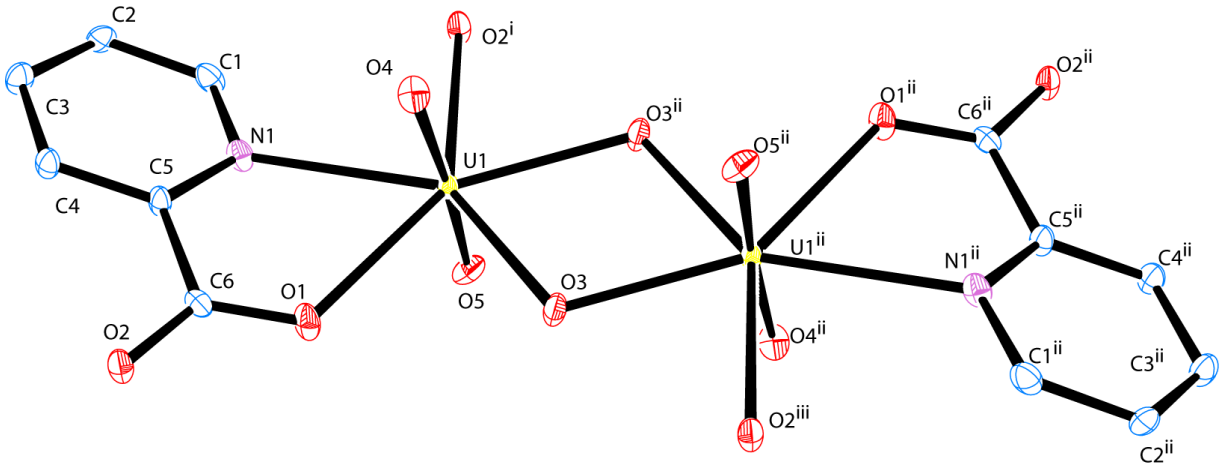


Figure S2. ORTEP illustration of **2**. Ellipsoids shown at 50% probability level. Superscript donates symmetry transformations i = x-1, y, z; ii = -x+1, y-2, z+1; iii = -x+2, -y+2,-z-1.

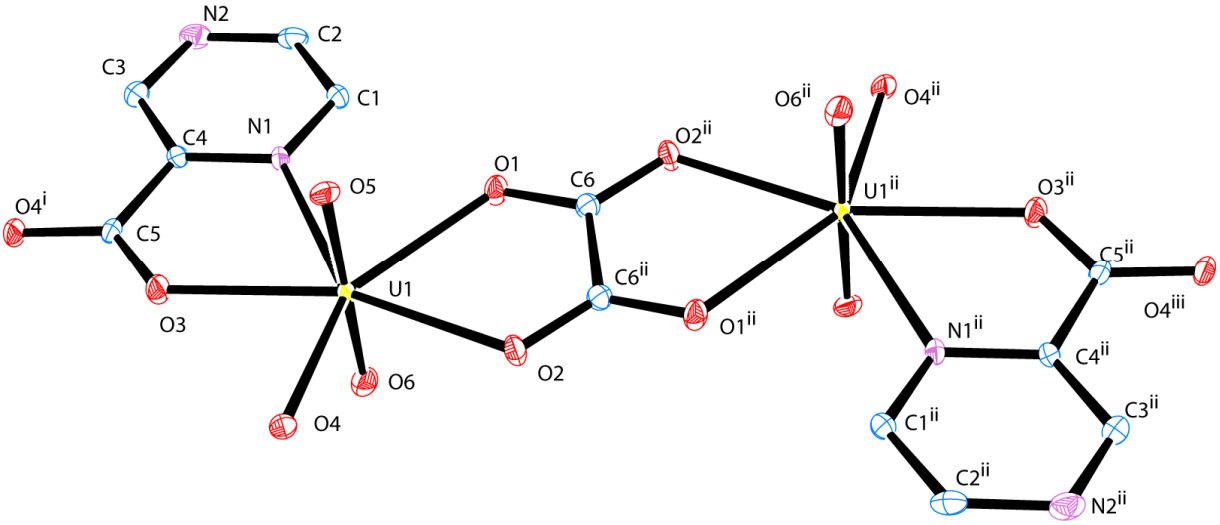


Figure S3. ORTEP illustration of **3**. Ellipsoids shown at 50% probability level. Superscript donates symmetry transformations i = -x+1, y-1/2, -z+3/2; ii = -x+2, -y+1, -z+2; iiix+1, -y+3/2, z+1/2.

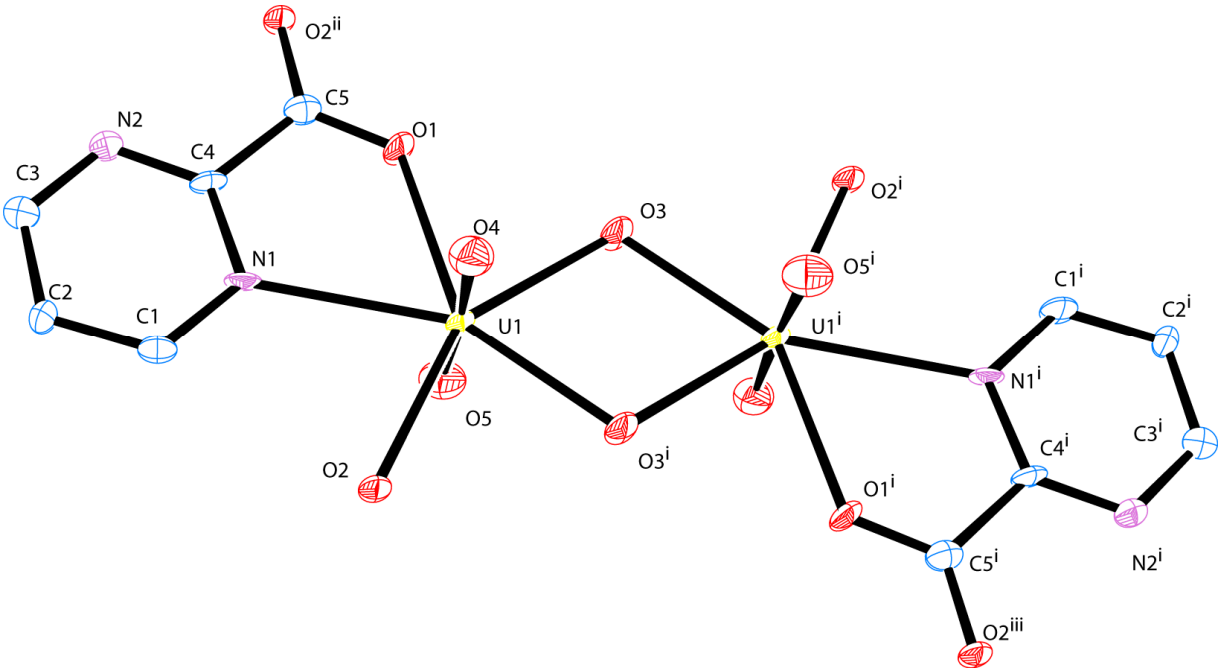


Figure S4. ORTEP illustration of **4**. Ellipsoids shown at 50% probability level. Superscript donates symmetry transformations i = -x+2, -y+1, -z+1; ii = x-1, y, z; iii = x+3, -y+1, -z+1.

Powder X-Ray Diffraction

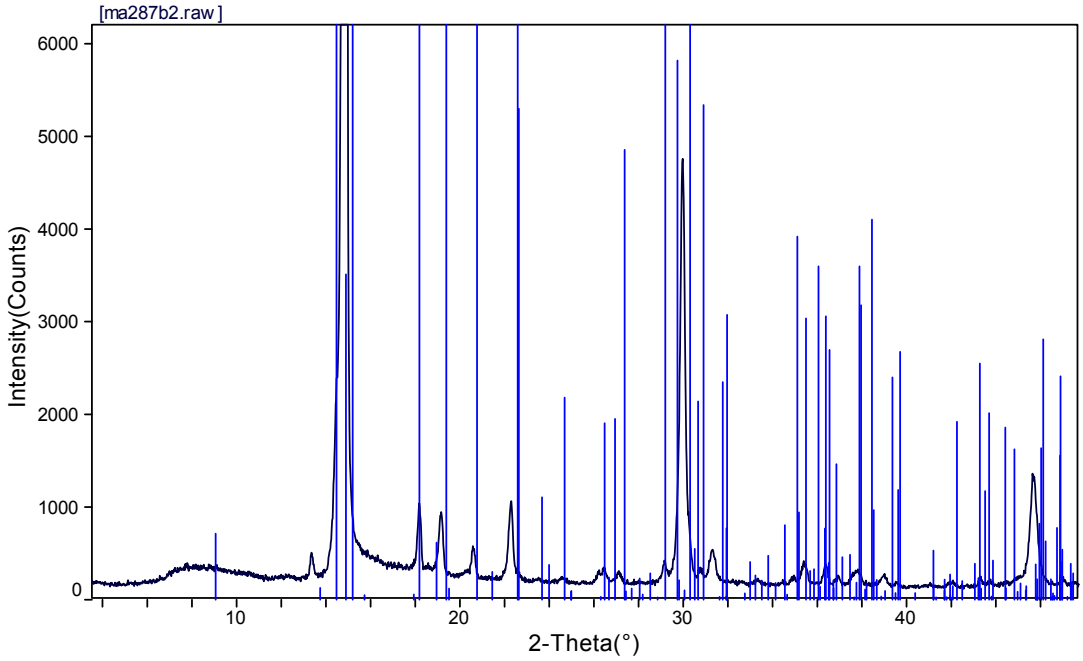


Figure S5. Observed PXRD pattern (black) for Compound **1** compared to calculated from CIF (blue)

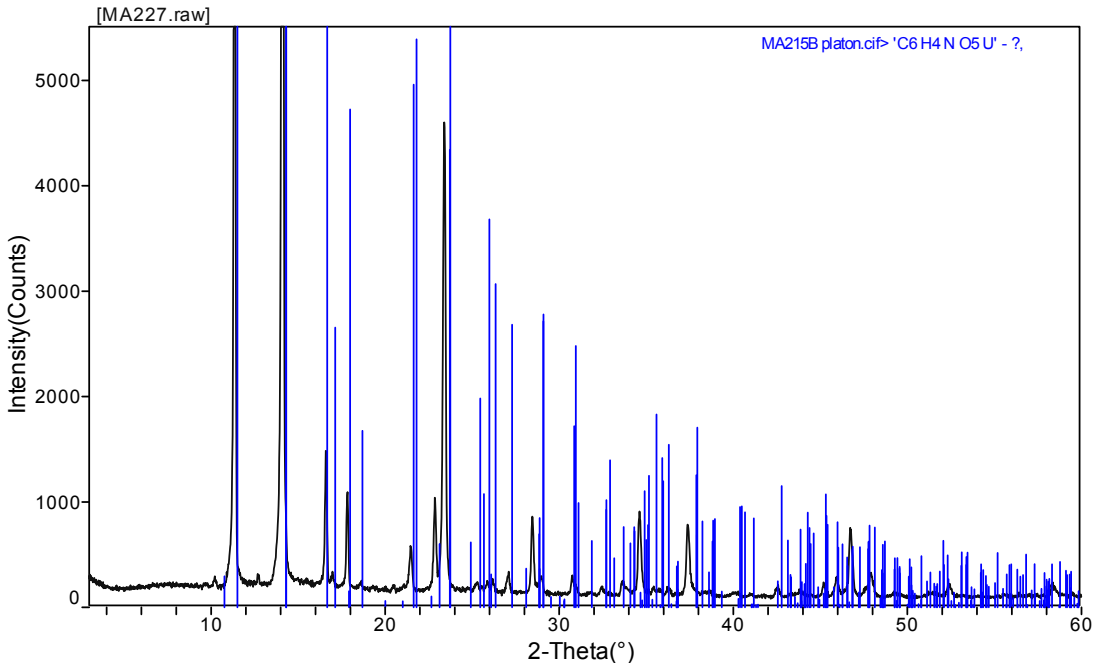


Figure S6. Observed PXRD pattern (black) for Compound **2** compared to calculated from CIF (blue)

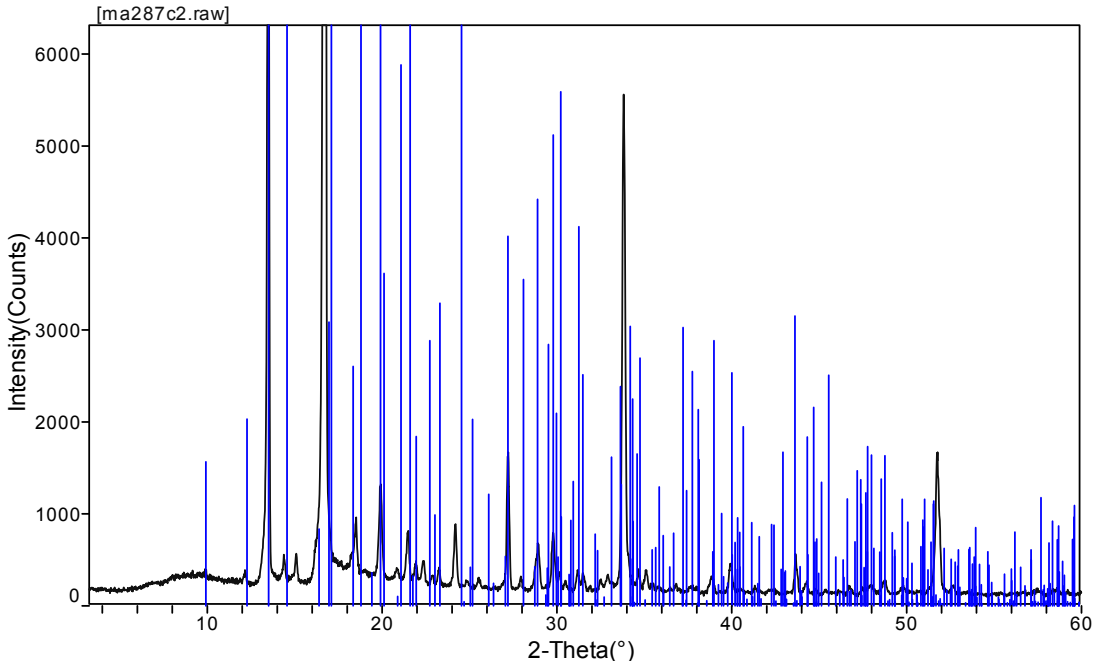


Figure S7. Observed PXRD pattern (black) for Compound **3** compared to calculated from CIF (blue)

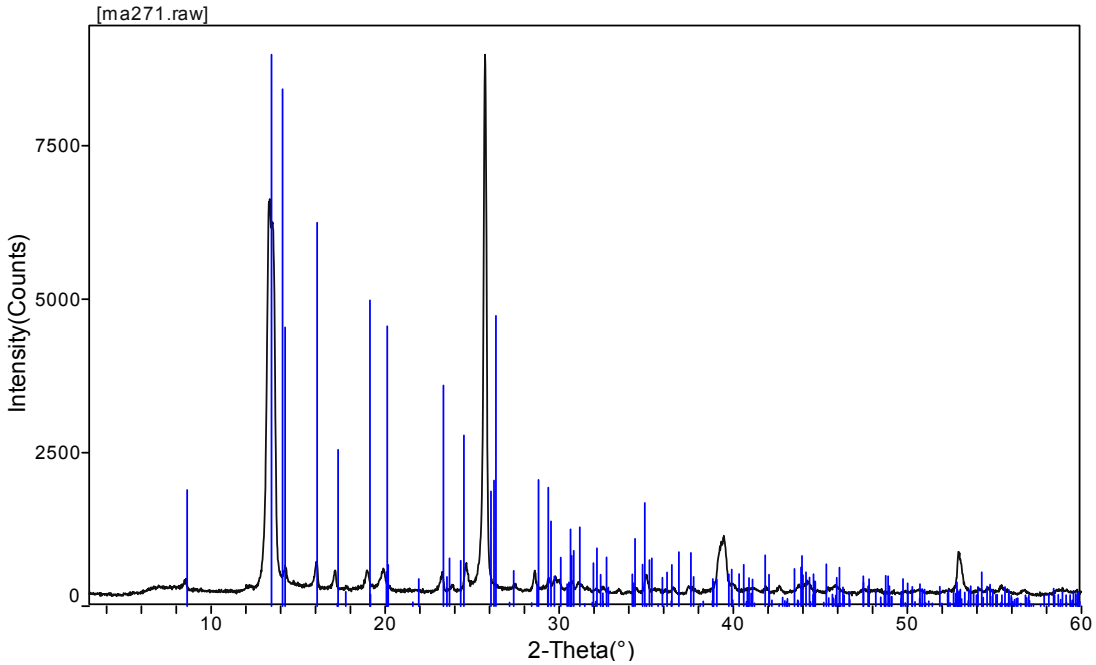


Figure S8. Observed PXRD pattern (black) for Compound **4** compared to calculated from CIF (blue)

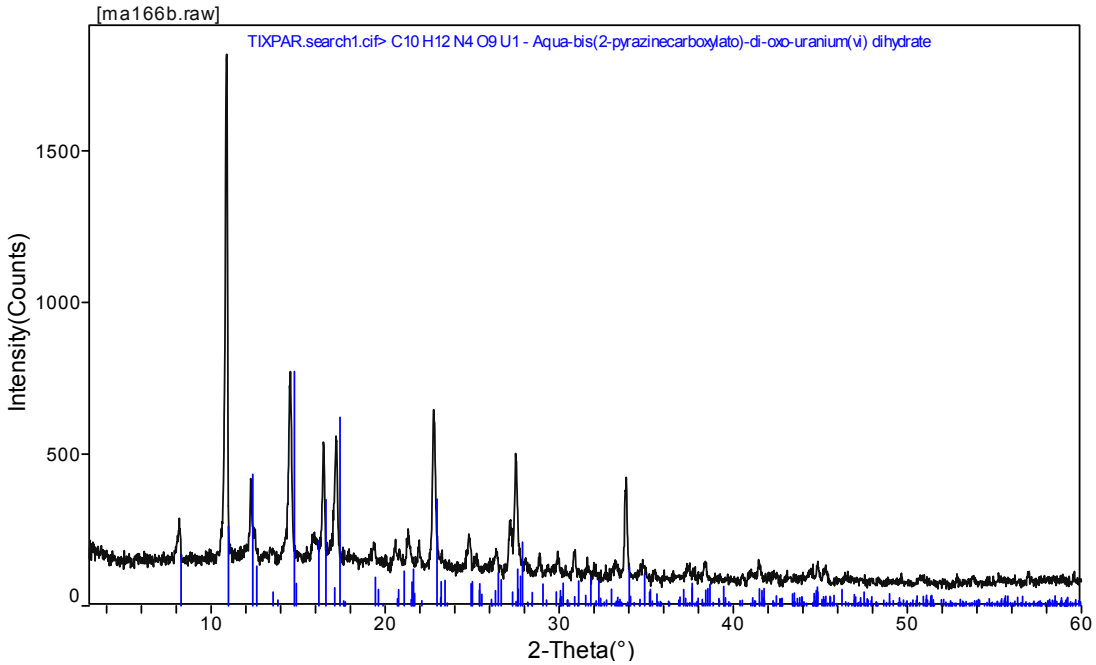


Figure S9. Observed PXRD pattern (black) for known uranyl pyrazine compound $[UO_2(C_5H_3N_2O_2)_2(H_2O)]·(H_2O)$[1] (CSD Refcode: TIXPAR) compared to calculated from CIF (blue).

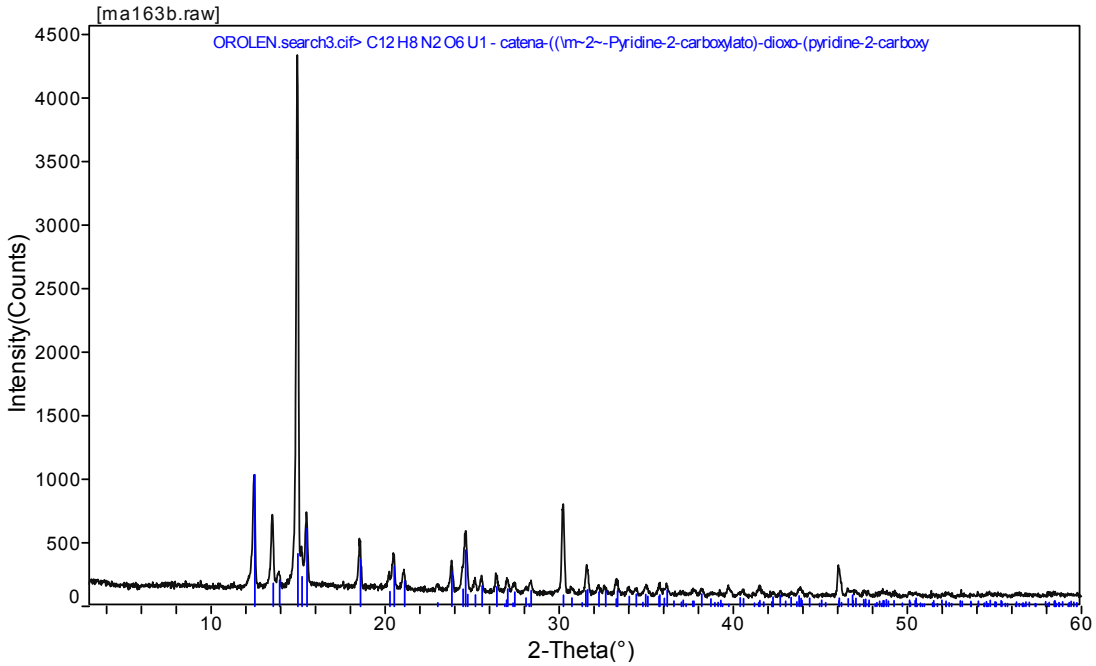


Figure S10. Observed PXRD pattern (black) for known uranyl pyridine material $[UO_2(C_6H_4N_1O_2)_2]$[2] compared to calculated from CIF (blue)

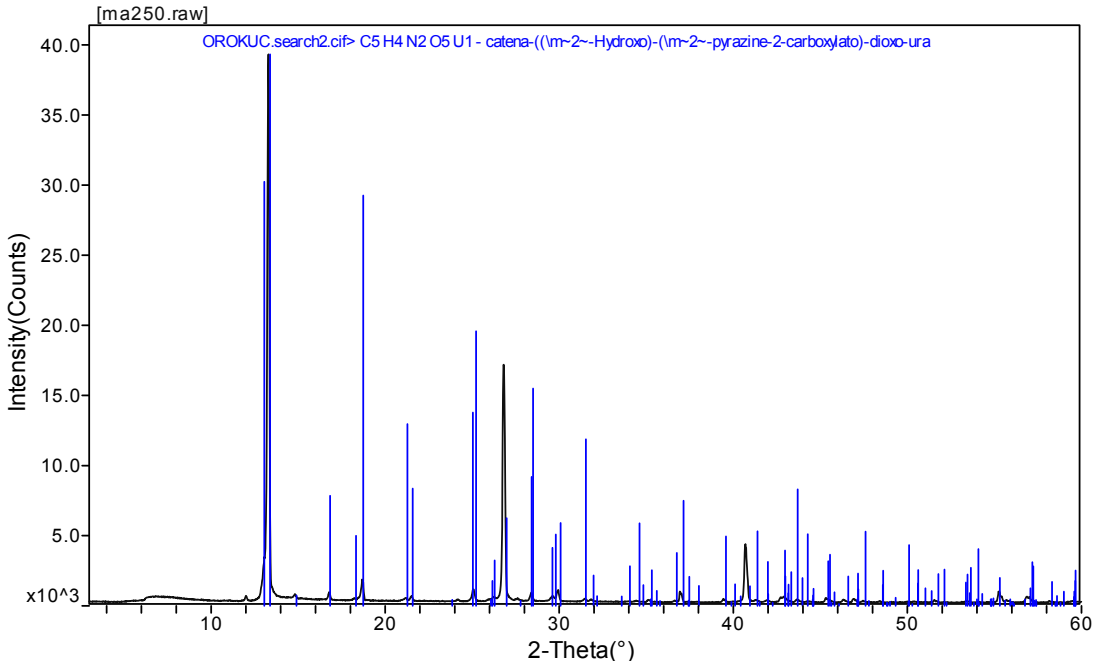


Figure S11. Observed PXRD pattern (black) for known uranyl pyrazine compound $[UO_2(C_5H_3N_2O_2)(OH)]^2$ compared to calculated from CIF (blue)

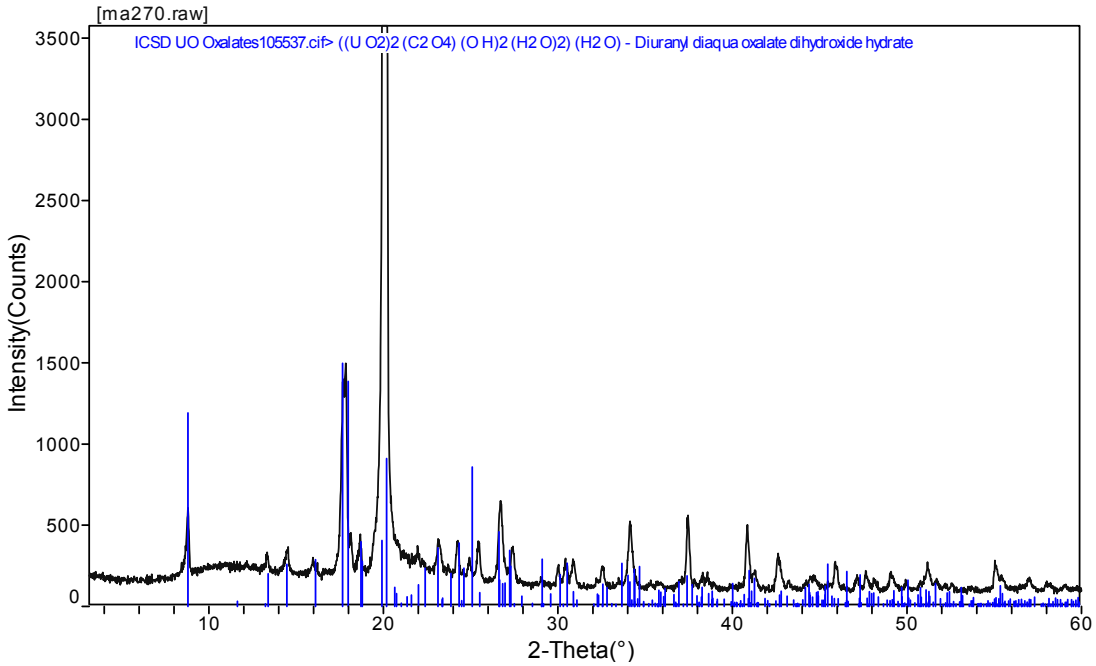


Figure S12. Observed PXRD pattern (black) for known uranyl oxalate compound $[(UO_2)_2(C_2O_4)(OH)_2(H_2O)]$□$(H_2O)$ (ICSD Refcode 105537).[3] compared to calculated from CIF (blue).

Thermogravimetric Analysis

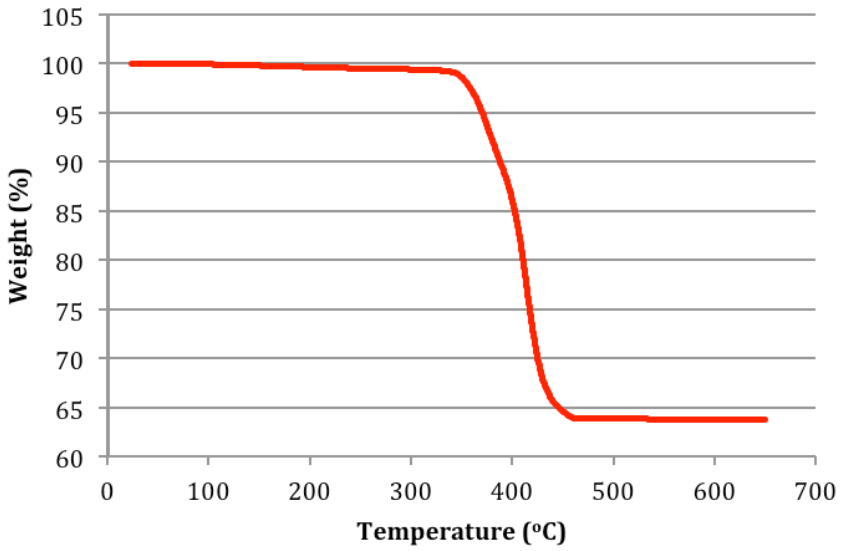
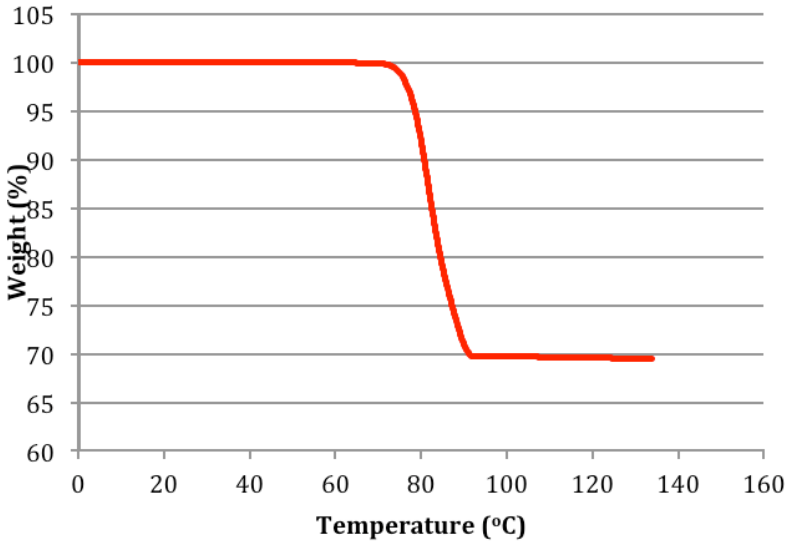
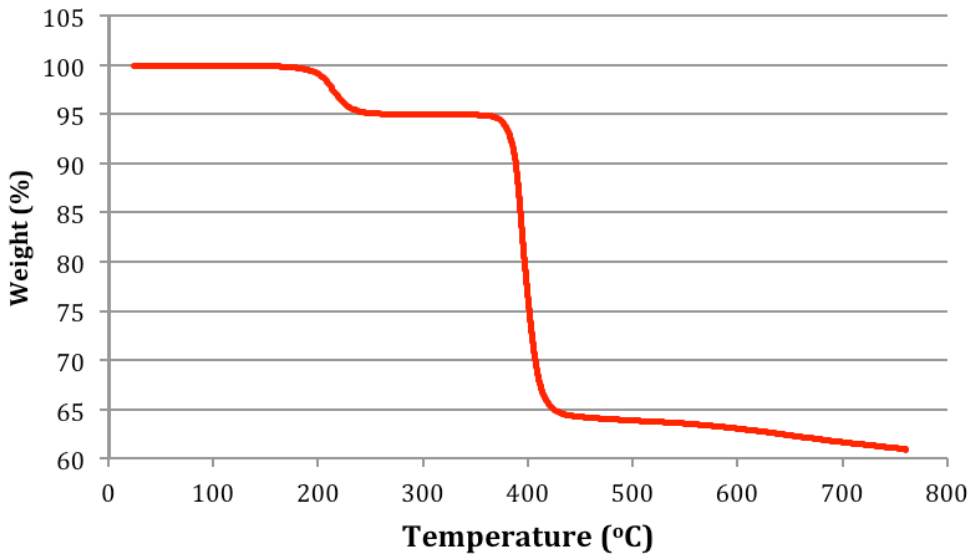


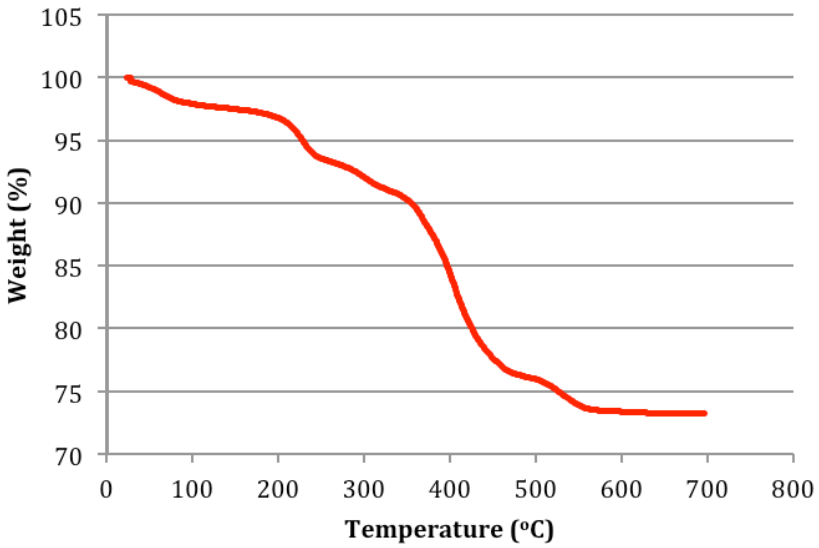
Figure S13. TGA of compound **1** showing 36% decrease in mass. Calculated mass of organic = 34%



SI Figure S14. TGA of compound **2** showing 30% decrease in mass. Calculated mass of organic = 30%



SI Figure S15. TGA of compound **3** showing 38% decrease in mass. Calculated mass of organic = 35%



SI Figure S16. TGA of compound **4** showing 28% decrease in mass. Calculated mass of organic = 31%

Elemental Analysis

Compound 1

ELEMENT	OBS	CALC
C	19.44 %	19.26 %
N	3.08 %	3.21 %
H	1.01 %	0.92 %

Compound 2

ELEMENT	OBS	CALC
C	17.80 %	17.60 %
N	3.31 %	3.42 %
H	1.31 %	1.22 %

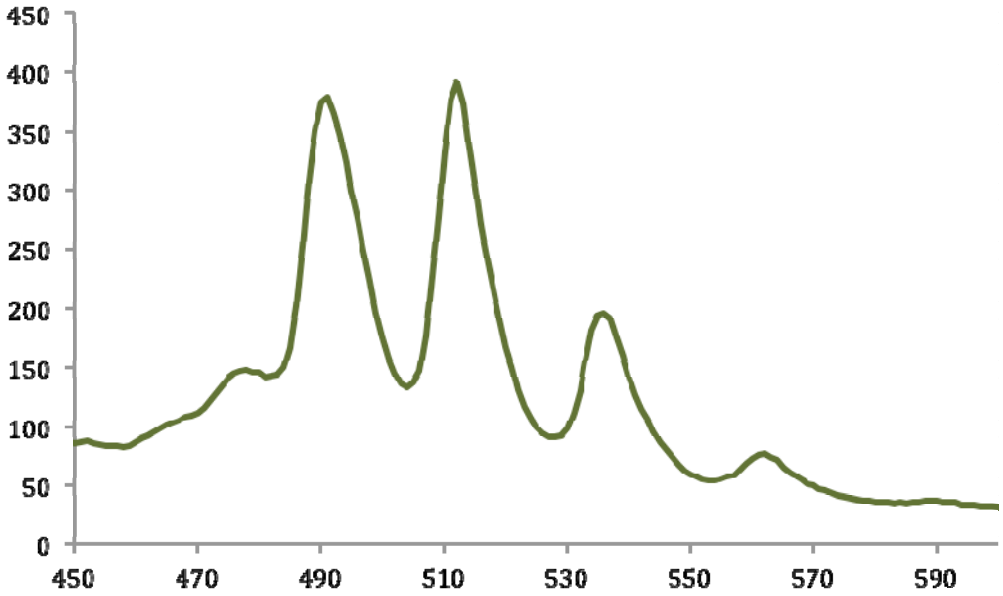
Compound 3

ELEMENT	OBS	CALC
C	15.99 %	15.89 %
N	6.08 %	6.18 %
H	1.18 %	0.66 %

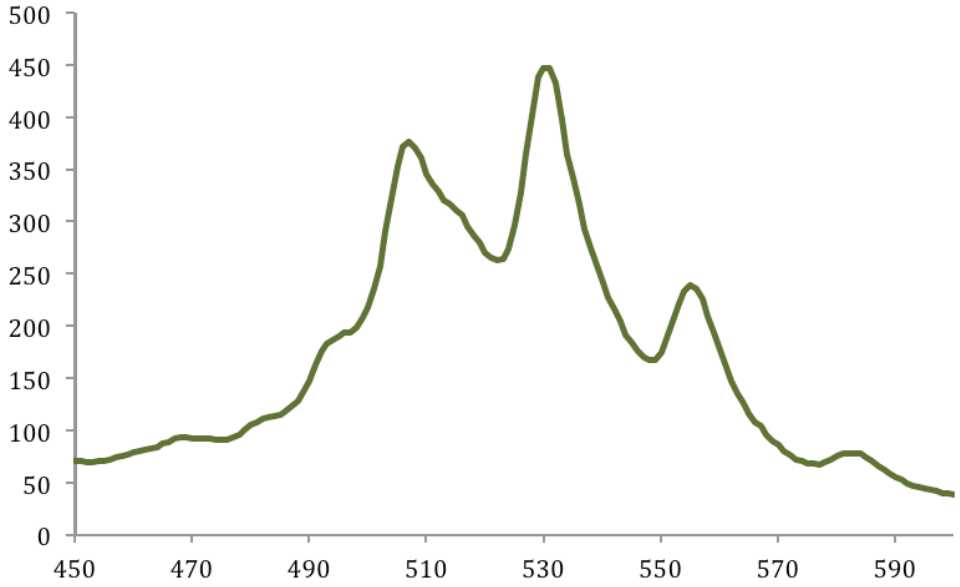
Compound 4

ELEMENT	OBS	CALC
C	15.12 %	14.08 %
N	6.11 %	6.61 %
H	1.42 %	0.93 %

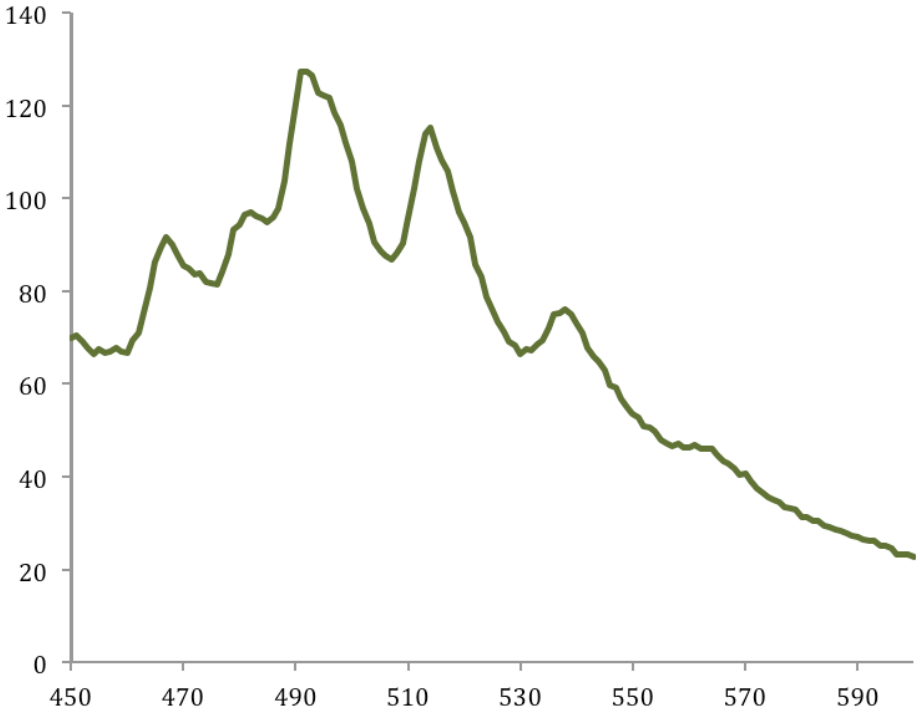
Fluorescence



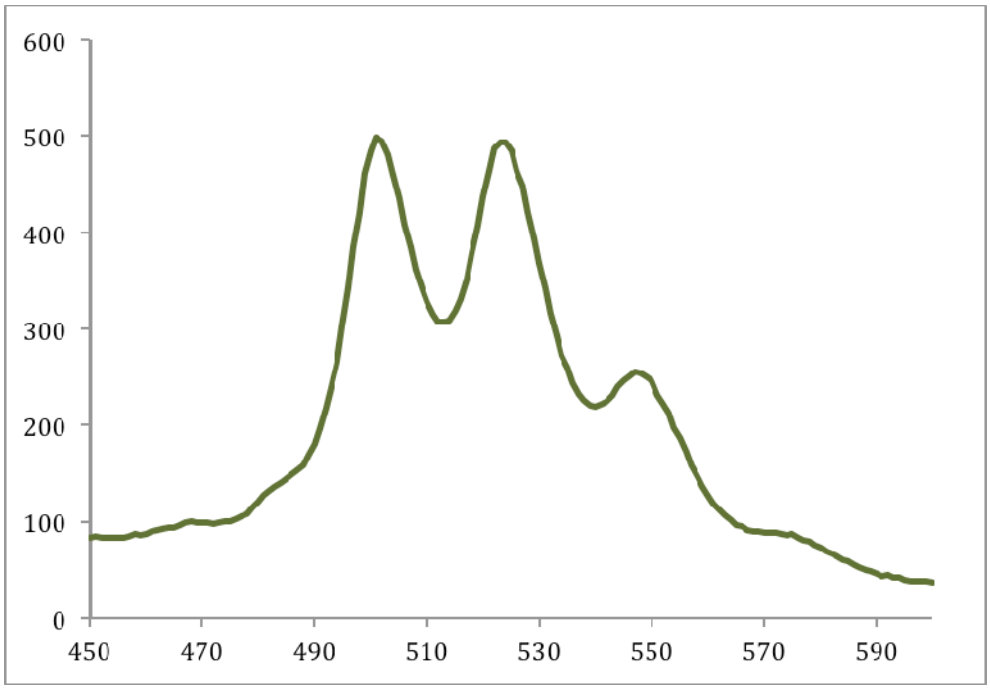
SI Figure 17. Fluorescence of compound **1**



SI Figure 18. Fluorescence of compound **2**



SI Figure 19. Fluorescence of compound **3**



SI Figure 20. Fluorescence of compound **4**

Bond valence calculation for bridging oxygen O3 in Compound 2[4, 5]

Atom	Bond Length	Valence Contribution
U1	2.306	0.604780212
U2	2.306	0.604780212
Total Valence		1.209560423

Bond valence calculation for bridging oxygen O3 in Compound 4[4, 5]

Atom	Bond Length	Valence Contribution
U1	2.333	0.574122038
U2	2.317	0.592097016
Total Valence		1.166219055

1. **N. W. Alcock, T. J. Kemp, S. M. Roe and J. Leciejewicz, *Inorganica Chimica Acta*, 1996, 248, 241-246.**
2. **R. C. Severance, S. A. Vaughn, M. D. Smith and H.-C. zur Loye, *Solid State Sciences*, 2011, 13, 1344-1353.**
3. **L. Duvieubourg, G. Nowogrocki, F. Abraham and S. Grandjean, *Journal of Solid State Chemistry*, 2005, 178, 3437-3444.**
4. **N. E. Brese and M. Okeeffe, *Acta Crystallographica Section B-Structural Science*, 1991, 47, 192-197.**
5. **P. C. Burns, R. C. Ewing and F. C. Hawthorne, *Canadian Mineralogist*, 1997, 35, 1551-1570.**