**Electronic Supporting Information**

# $PF_6^-$ Hydrolysis as a Route to Unique Uranium Phosphate Materials

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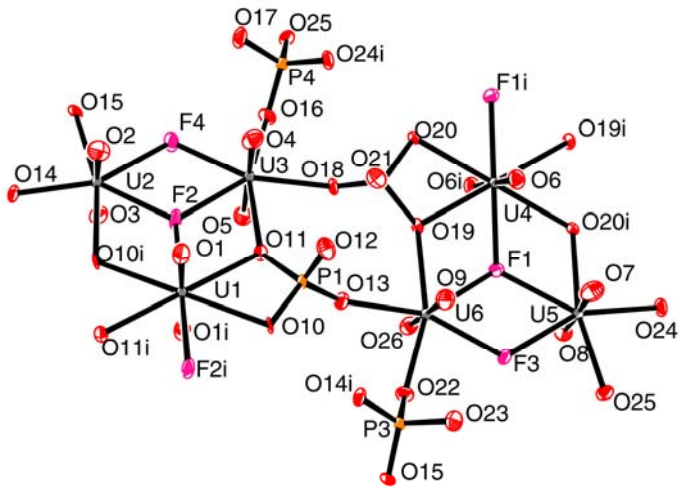


**Fig S1**. ORTEP illustration of **1**. Ellipsoids are shown at 50% probability level. Inter-layer organic molecules are omitted for clarity. Superscript denotes symmetry transformations i = -x, -y, -z.

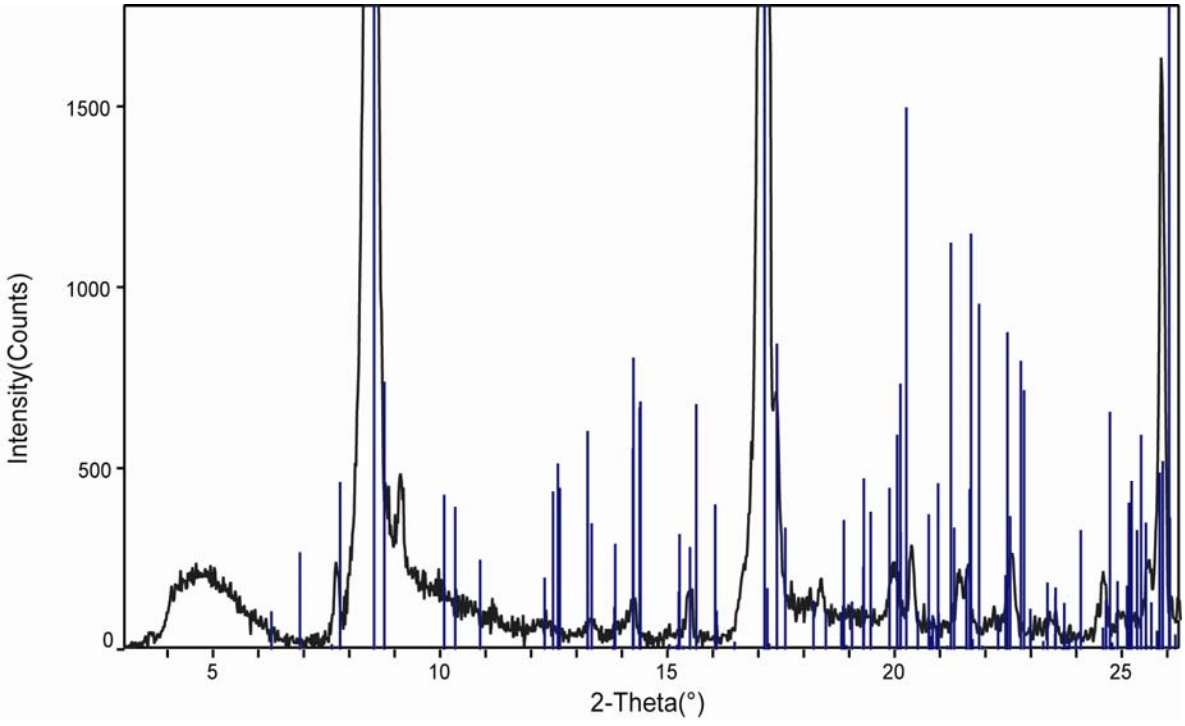


**Fig S2**. Powder diffraction spectra of **1** with calculated pattern overlaid in blue. We acknowledge a minor impurity peak at ~2θ = 9.1º.

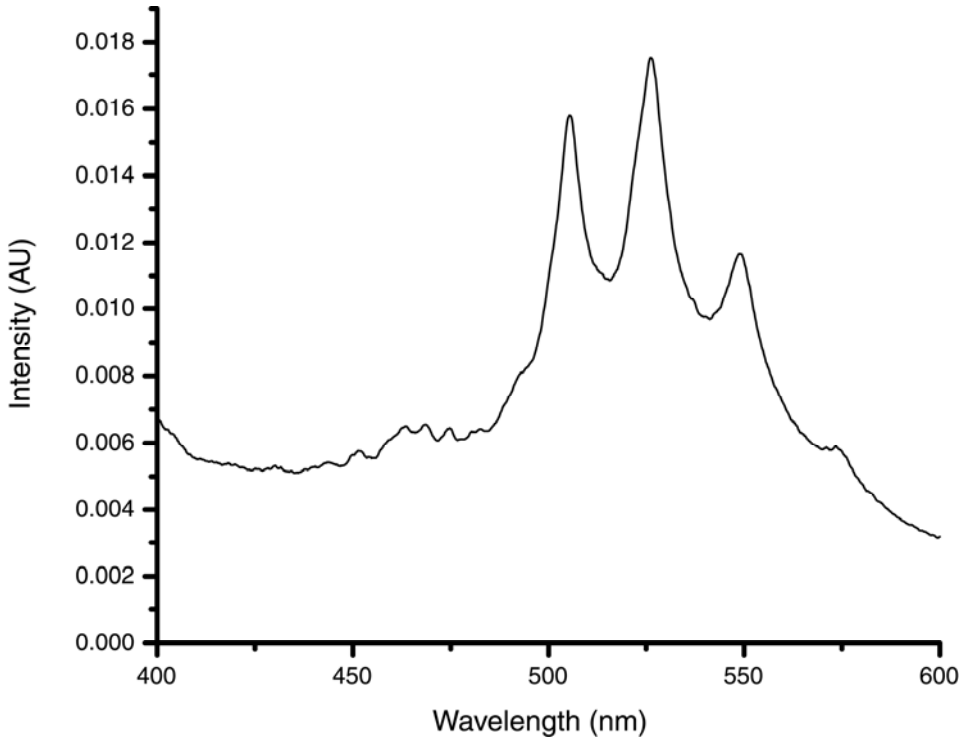


**Fig S3**. Emission spectra for compound **1** via direct uranyl excitation at 365 nm.

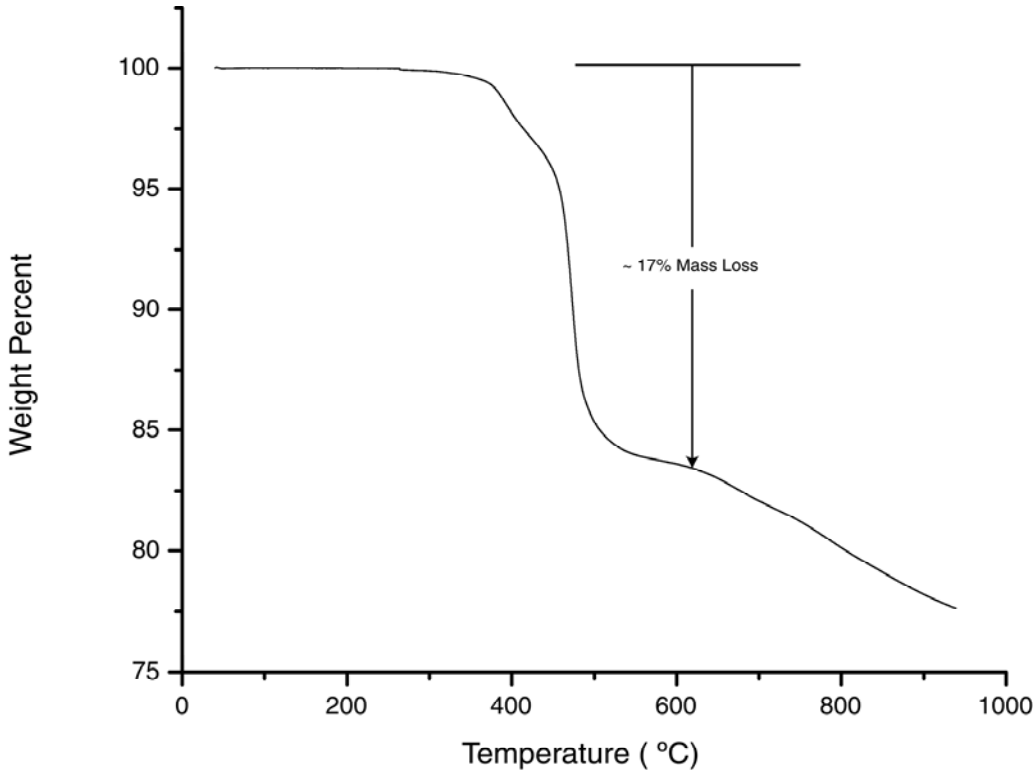


**Fig S4**. Thermogravametric analysis of **1** in flowing $N_2$.

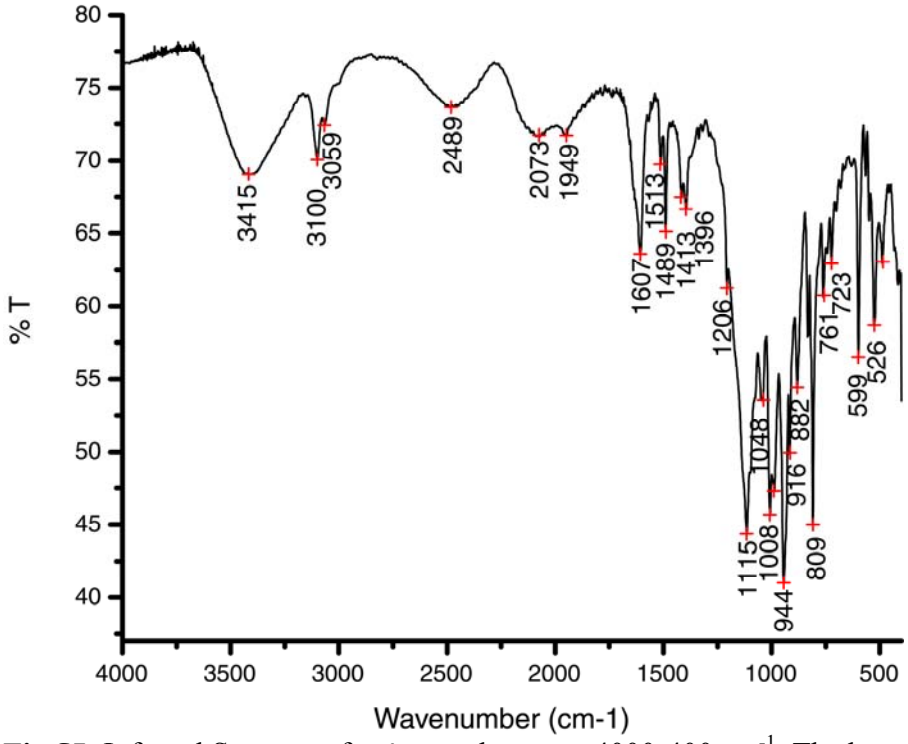


**Fig S5**. Infrared Spectrum for **1** over the range 4000-400 $cm^{-1}$. The broad stretch centered at 3415 $cm^{-1}$ consistent with protonated bpy, and the strong stretch at 1115 $cm^{-1}$ consistent with the P=O bonds of the phosphates. C-H stretches of the organic are observed at 3100 and 3059 $cm^{-1}$.

**Table S1**. Bond valence sums calculated using the parameters of Burns et al. (1997) for $U^{6+}$and non-uranyl O, and Brese and O'Keeffe(1991) for F.[1, 2]

| | | | |
|---|---|---|---|
| U1 | 6.09 | O13 | 1.86 |
| U2 | 6.00 | O14 | 1.87 |
| U3 | 5.97 | O15 | 1.87 |
| U4 | 6.00 | O16 | 1.83 |
| U5 | 5.97 | O17 | 1.05 |
| U6 | 6.01 | O18 | 1.89 |
| F1 | 1.26 | O19 | 2.06 |
| F2 | 1.32 | O20 | 2.03 |
| F3 | 0.86 | O21 | 1.25 |
| F4 | 0.89 | O22 | 1.87 |
| O10 | 2.05 | O23 | 1.02 |
| O11 | 2.11 | O24 | 1.86 |
| O12 | 1.21 | O25 | 1.86 |

1. N. E. Brese and M. O'Keeffe, *Acta Crystallog., Sect. B: Struct. Sci.*, 1991, 192-197.
2. P. C. Burns, *Can. Mineral.*, 1997, **35**, 1551-1570.