Electronic Supplementary Material (ESI) for Dalton Transactions.

Supplementary Information for
Incorporation of Iodine into Uranium Oxyhydroxide Phases

Gabriel L. Murphy,[1*] Philip Kegler,[1] Martina Klinkenberg,[1] Andreas Wilden,[1] Maximilian Henkes,[1] Dimitri Schneider,[1] and Evgeny V. Alekseev[1*]

[1]*Institute of Energy and Climate Research, Forschungszentrum Jülich GmbH, 52428 Jülich, Germany*

Corresponding authors: Gabriel L. Murphy and Evgeny V. Alekseev

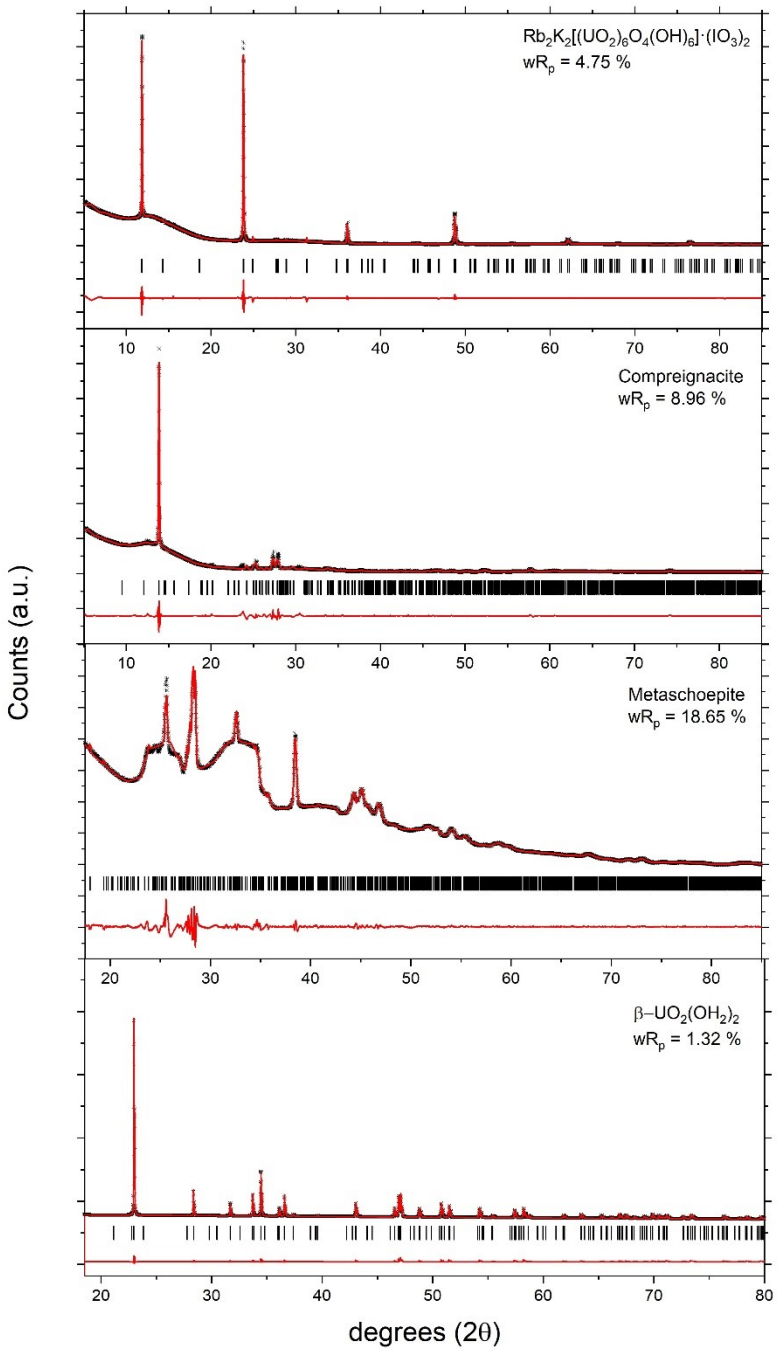
Table of Contents

Section S1: Powder X-ray Diffraction and Analysis

XRD data collected against samples of $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$, metaschoepite, compreignacite and β-$UO_2(OH)_2$ were analysed using profile matching in the program Fullprof.[1] A starting model using the lattice parameters determined from either SC-XRD measurements given previously or from reference cif files[2] were used and refined with the zero point, peak shape (pseudo-Voigt function) and instrument parameters against the collected data. The refinement profiles are given in Supplementary Information Figure S2 where it can be observed all samples are single phase. Supplementary Information Table S3 provides the refined lattice parameters and compares them against the SC-XRD solution value for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ and against metaschoepite, compreignacite and β-$UO_2(OH)_2$ references.

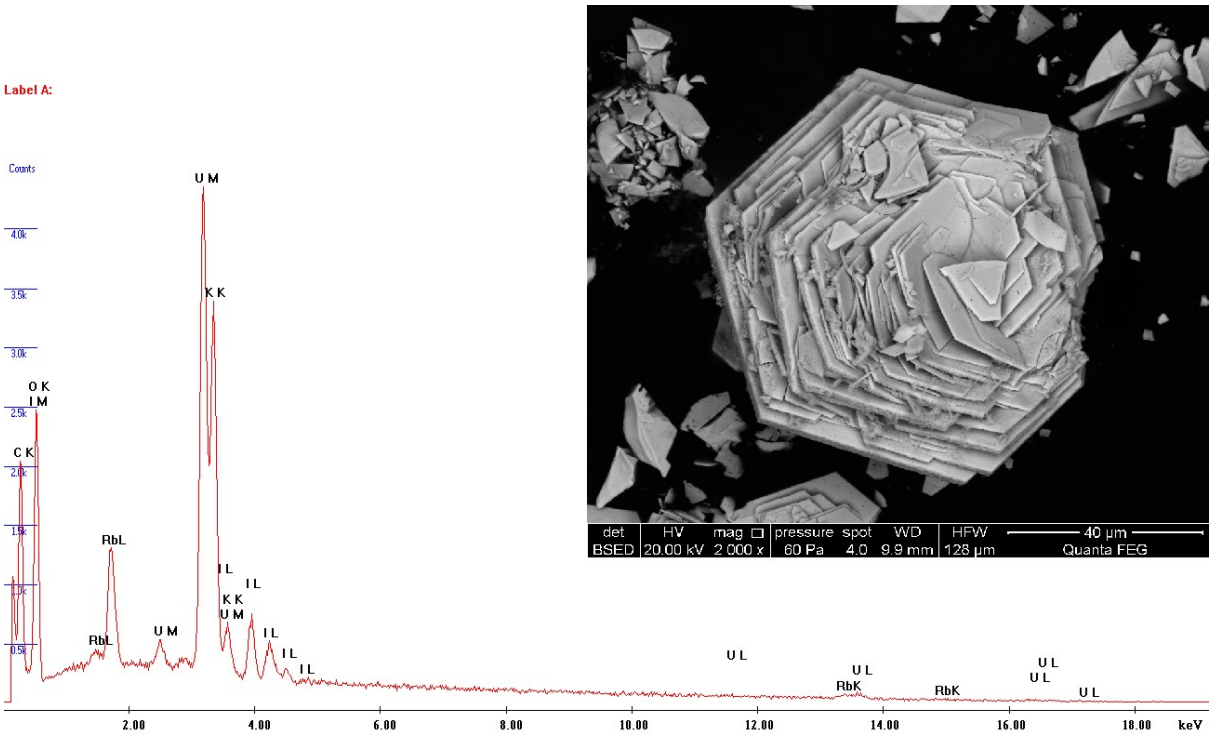
Supplementary Information Table S1. Refined lattice parameters for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$, metaschoepite, compreignacite and β-$UO_2(OH)_2$ derived from XRD measurements compared against reference values.

Phase	**Chemical Formula**	**Study**	**Structure**	**Space group**	***a* (Å)**	***b* (Å)**	***c* (Å)**	**α (°)**	**β (°)**	**γ (°)**	**wR_p**
$Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$	$Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$	Present	Trigonal	$P3_1m$	7.2069(17)	7.2069(17)	7.510(3)	90	90	120	4.75%
Compreignacite	$K_2[(UO_2)_6O_4(OH)_6]\cdot 7H_2O$	Present	Orthorhombic	*Pnnm*	14.8759(5)	7.1738(4)	12.2249(8)	90	90	90	8.96%
"	"	Burns *et al.*[3]	Orthorhombic	*Pnnm*	14.8591(7)	7.1747(3)	12.1871(5)	90	90	90	-
Metaschoepite	$[(UO_2)_8O_2(OH)_{12}]\cdot 12H_2O$	Present	Orthorhombic	*Pbcn*	14.271(6)	16.808(7)	14.902(6)	90	90	90	18.65%
"	"	Weller *et al.*[4]	Orthorhombic	*Pbcn*	14.050(2)	16.709(2)	14.7291(2)	90	90	90	-
β-$UO_2(OH)_2$	β-$UO_2(OH)_2$	Present	Orthorhombic	*Pbca*	5.6443(2)	6.2884(2)	9.9366(4)	90	90	90	1.32 %
"	"	Taylor & Hurst[5]	Orthorhombic	*Pbca*	5.6438(1)	6.2871(1)	9.9372(2)	90	90	90	-



Supplementary Information Figure S1. XRD and profile matching analysis profiles using FullProf for (top to bottom) $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$, compreignacite, metaschoepite and β-$UO_2(OH)_2$. Note the low angle humps present in) $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$, compreignacite, metaschoepite are related to the dome enclosure used for sample measurement due to the radioactive content.

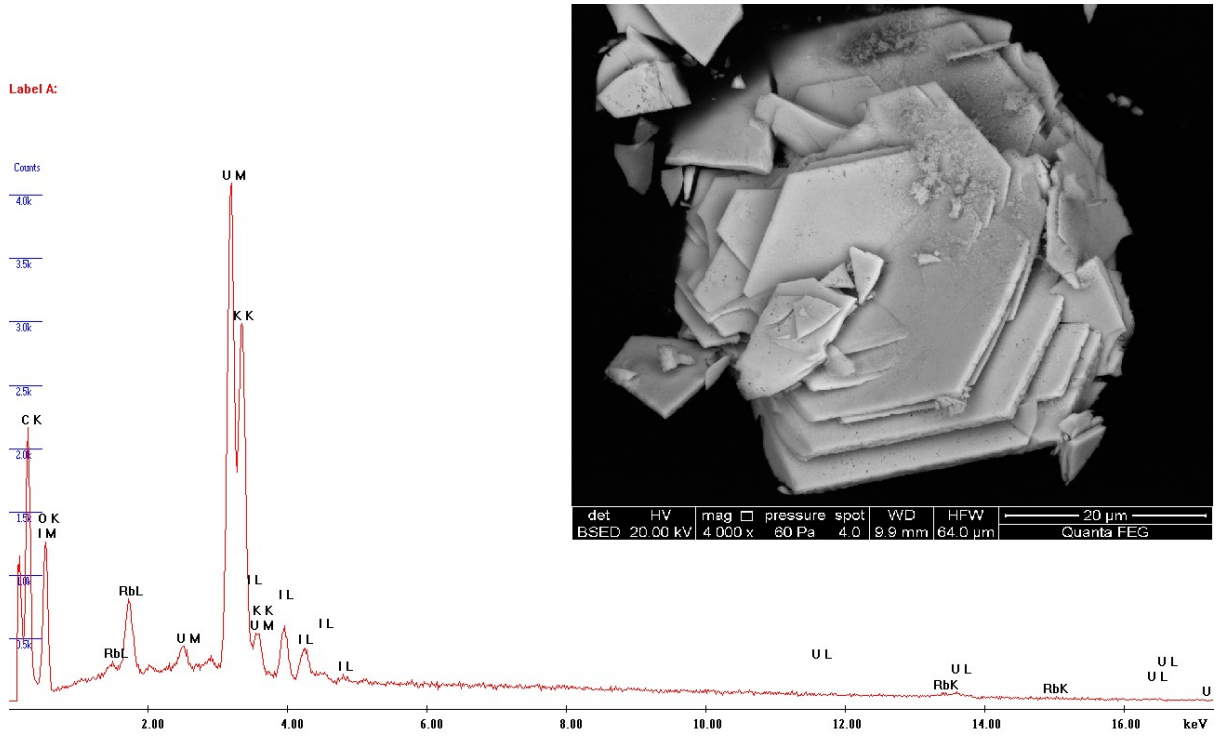
Section S2: Scanning Electron Microscopy



Supplementary Information Figure S2a. SEM micrograph for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

Supplementary Information Table S3a. Normalised (to U content) EDS result for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

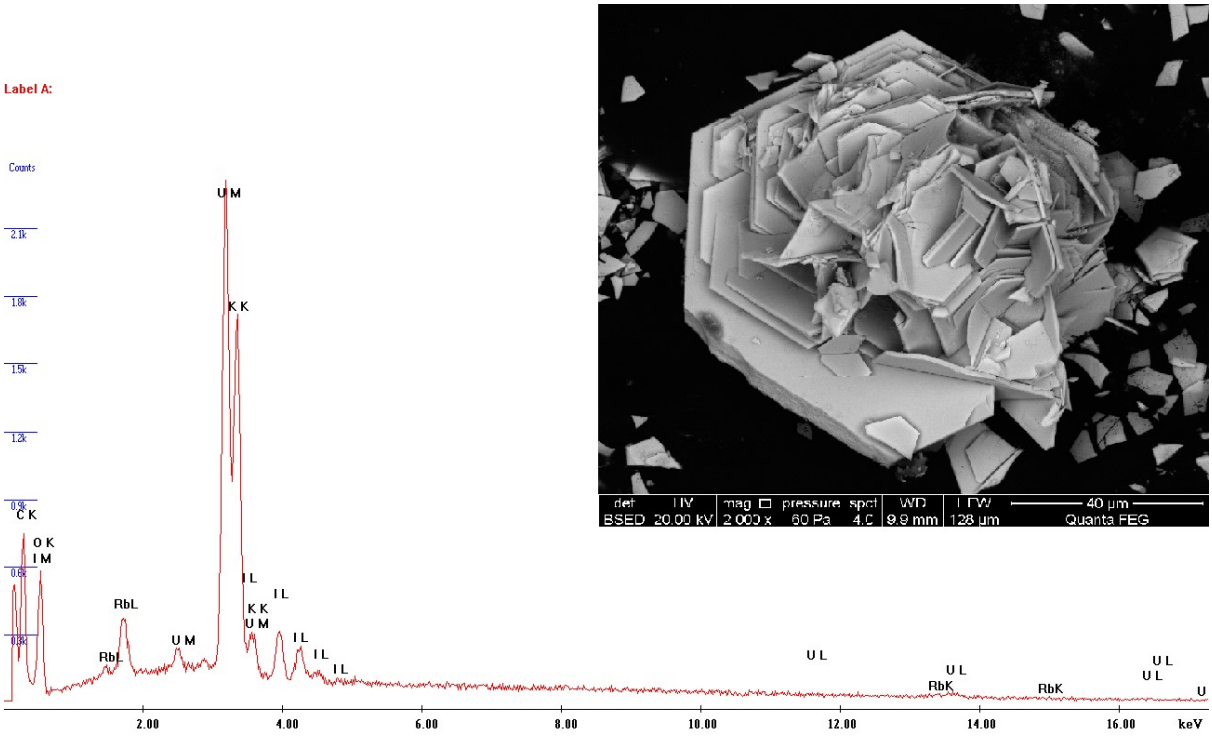
Element	U	I	K	Rb
Total	1	0.30	0.35	0.44



Supplementary Information Figure S2b. SEM micrograph for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

Supplementary Information Table S3b. Normalised (to U content) EDS result for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

Element	U	I	K	Rb
Total	1	0.25	0.38	0.24



Supplementary Information Figure S2c. SEM micrograph for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

Supplementary Information Table S3c. Normalised (to U content) EDS result for $Rb_2K_2[(UO_2)_6O_4(OH)_6]\cdot(IO_3)_2$ crystal specimen.

Element	U	I	K	Rb
Total	1	0.28	0.33	0.26

Section S3: References

1. Rodríguez-Carvajal, J. Recent advances in magnetic structure determination by neutron powder diffraction. *Physica B: Condensed Matter* **1993,** 192 (1), 55-69 DOI: https://doi.org/10.1016/0921-4526(93)90108-I.
2. Gorman-Lewis, D.; Fein, J. B.; Burns, P. C.; Szymanowski, J. E.; Converse, J. Solubility measurements of the uranyl oxide hydrate phases metaschoepite, compreignacite, Na–compreignacite, becquerelite, and clarkeite. *The Journal of Chemical Thermodynamics* **2008,** 40 (6), 980-990.
3. Burns, P. C. The structure of compreignacite, K 2 (UO 2) 3 O 2 (OH) 3] 2 (H 2 O) 7. *The Canadian Mineralogist* **1998,** 36 (4), 1061-1067.
4. Weller, M. T.; Light, M. E.; Gelbrich, T. Structure of uranium(VI) oxide dihydrate, UO3.2H2O; synthetic meta-schoepite (UO2)4O(OH)6.5H2O. *Acta Crystallographica Section B* **2000,** 56 (4), 577-583 DOI: doi:10.1107/S0108768199016559.
5. Taylor, J. C.; Hurst, H. J. The hydrogen-atom locations in the [alpha] and [beta] forms of uranyl hydroxide. *Acta Crystallographica Section B* **1971,** 27 (10), 2018-2022 DOI: doi:10.1107/S0567740871005259.