

Electronic Supplementary Information

Investigation of uranium oxide hydrates with barium(II) ions:
structural diversity, uranium valences, and implications to uranium
geochemistry and spent nuclear fuel [†]

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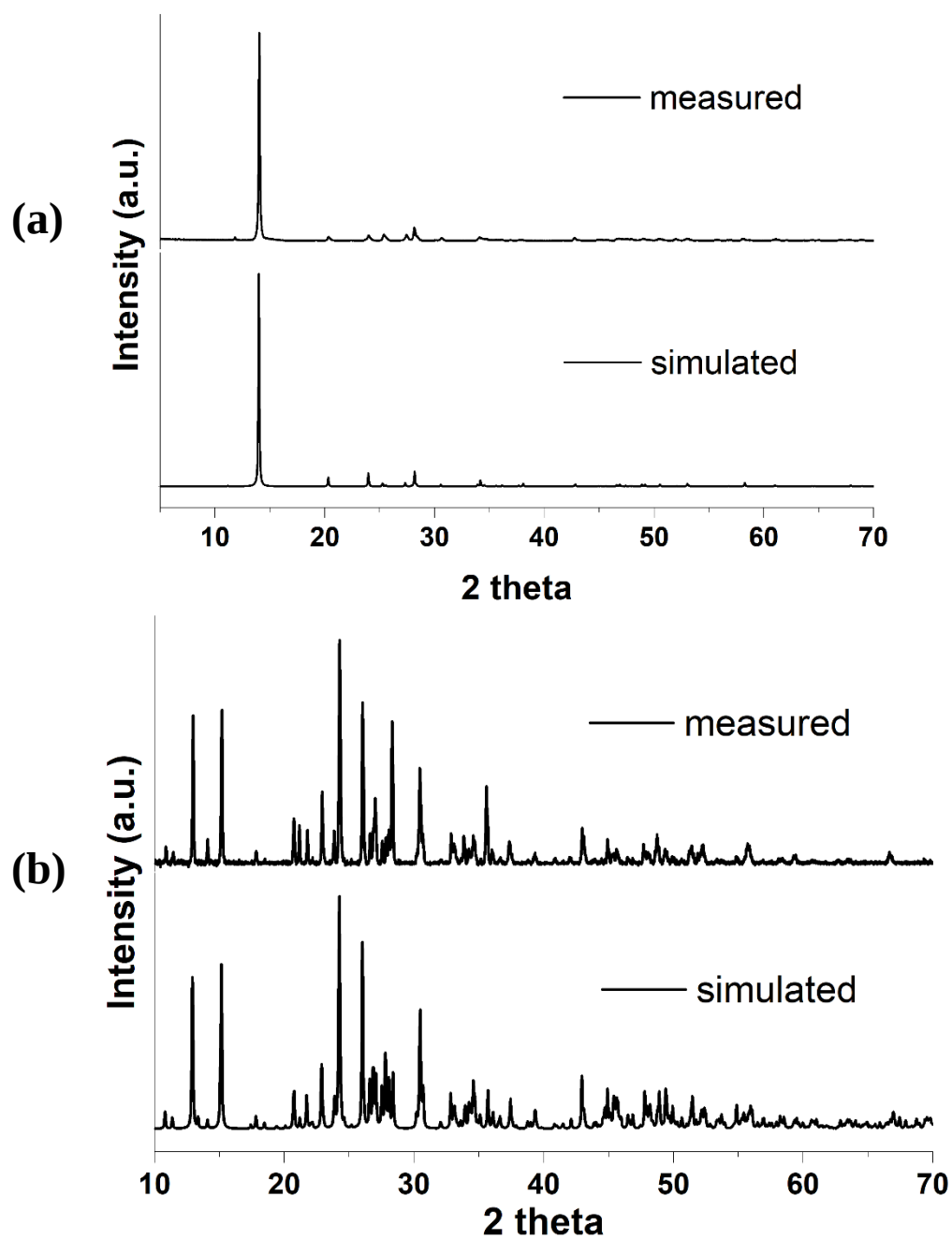


Fig. S1. Powder XRD patterns of compounds **UOH-Ba1** (a) and **UOH-Ba2** (b) with measured on top of the simulated ones.

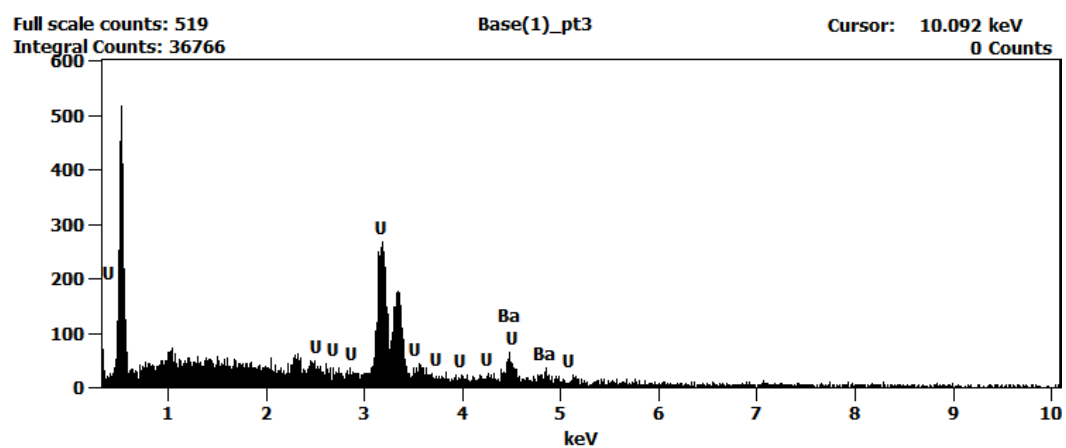
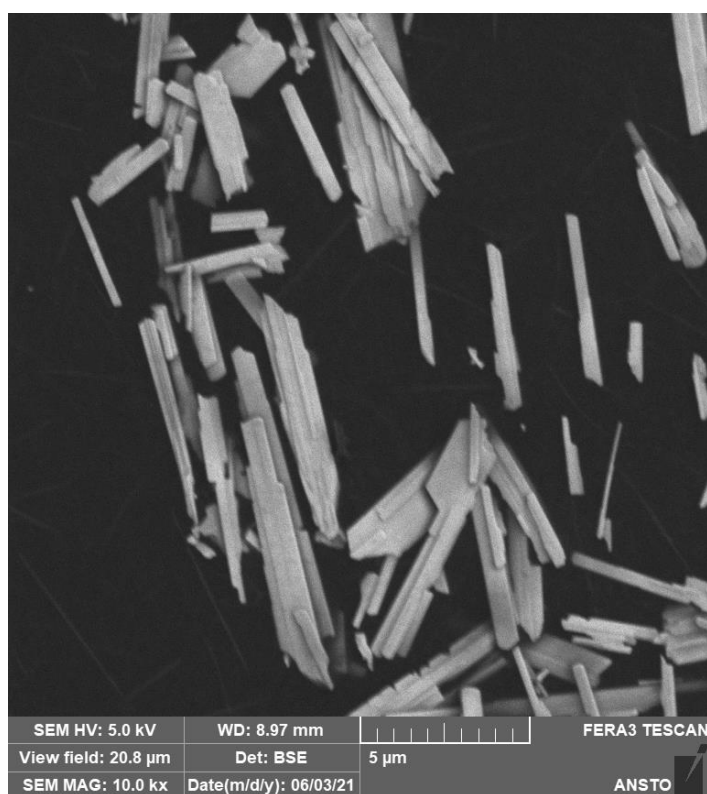


Fig. S2. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound **UOH-Ba1** prepared with U_3O_8 as the uranium precursor.

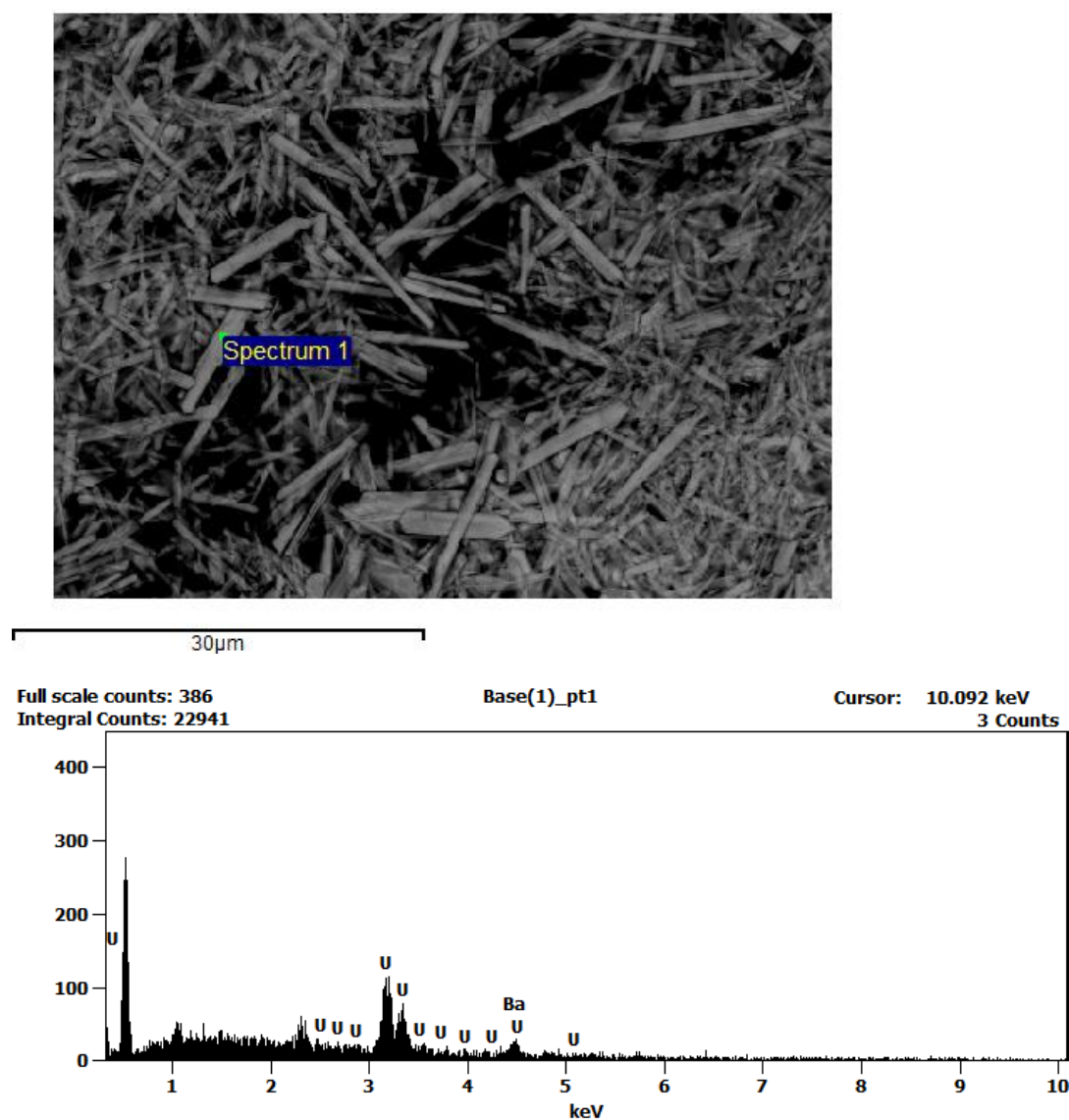


Fig. S3. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound **UOH-Ba1s** prepared with schoepite as the uranium precursor.

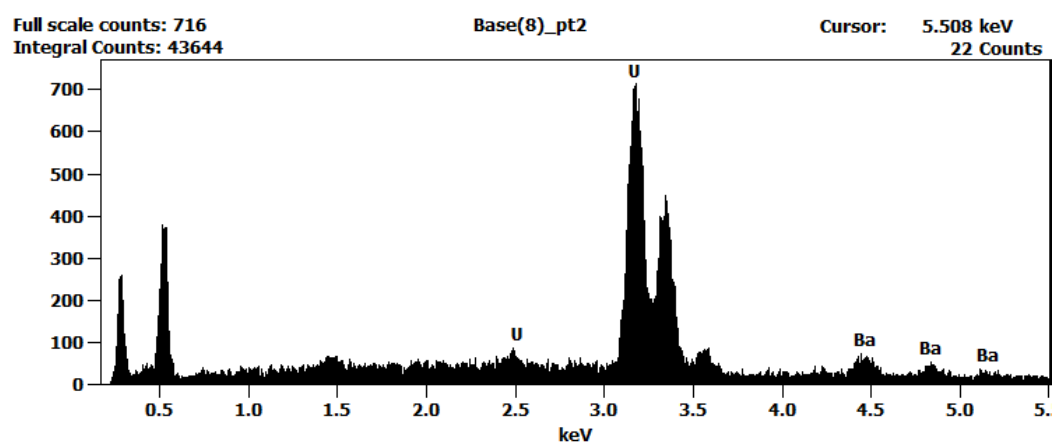
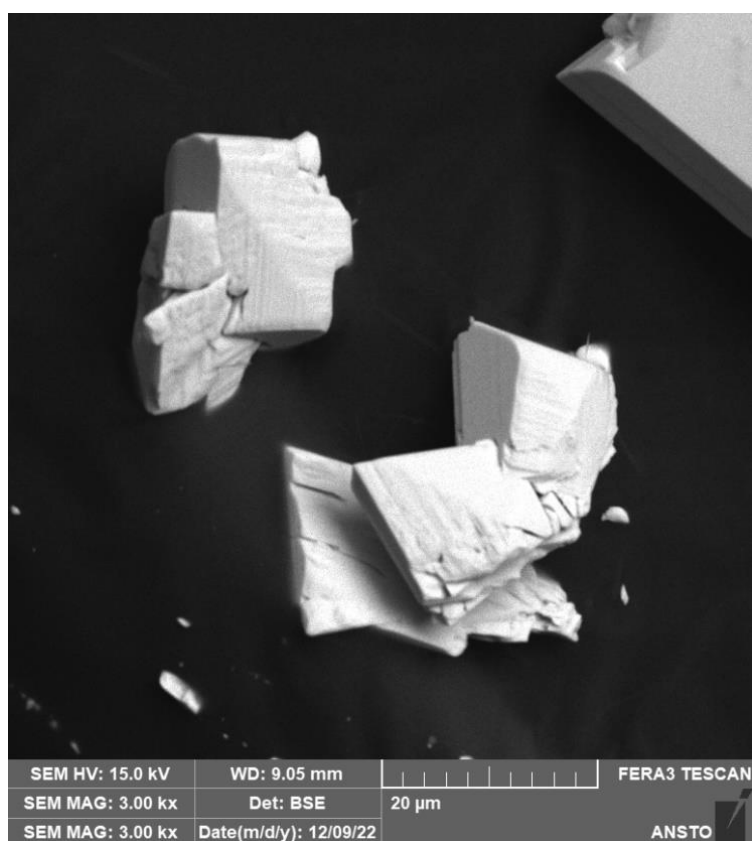


Fig. S4. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound UOH-Ba₂.

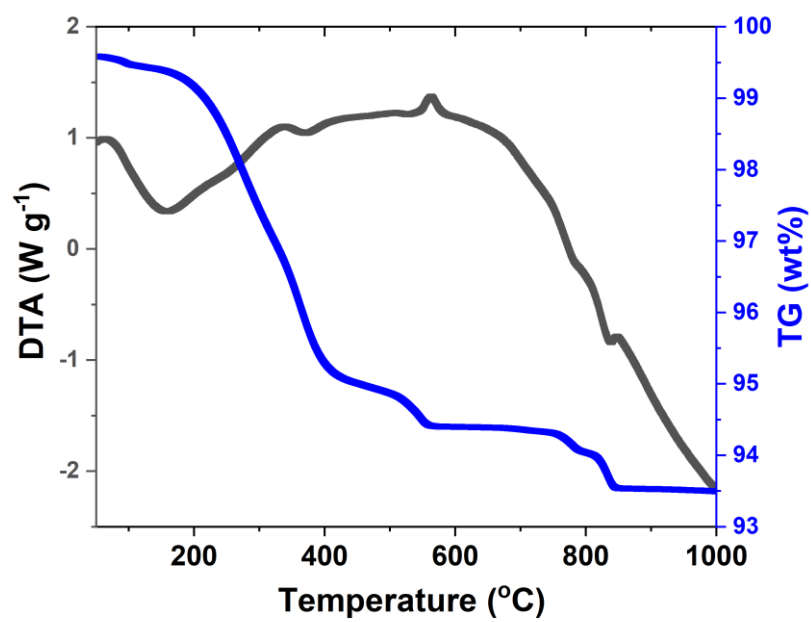


Fig. S5. Thermogravimetric analysis results for compound **UOH-Ba1**.

Table S1. Selected bond lengths and angles for compound **UOH-Ba1**.

Bond	Length (Å)	Bond	Length (Å)
U1-O1	1.77(3)	U2-O4	1.77(4)
U1-O2	1.78(3)	U2-O5	1.79(4)
U1-O3 ²	2.31(4)	U2-O6 ⁷	2.27(3)
U1-O3	2.23(3)	U2-O6	2.37(3)
U1-O3 ³	2.28(4)	U2-O6 ⁶	2.22(3)
U1-O7 ⁴	2.37(4)	U2-O7	2.32(5)
U1-O8 ⁵	2.21(4)	U2-O8	2.20(4)
O1=U1=O2	177.3(17)	O4=U2=O5	176.3(18)
Ba-O1 ²	3.06(4)	Ba-O4 ⁶	2.80(5)
Ba-O1 ⁹	2.77(3)	Ba-O5	3.08(3)
Ba-O2 ¹⁰	2.76(3)	Ba-O6	3.29(3)
Ba-O2	2.72(3)	Ba-O9 ³	2.869(5)
Ba-O3	3.26(4)	Ba-O9	2.865(5)
Ba-O4 ⁷	2.78(6)		

¹2-X,2-Y,-Z; ²1-X,2-Y,-Z; ³1+X,+Y,+Z; ⁴-X,2-Y,1-Z; ⁵1-X,2-Y,1-Z; ⁶1-X,1-Y,1-Z; ⁷-X,1-Y,1-Z; ⁸1-X,1-Y,-Z; ⁹+X,-1+Y,+Z; ¹⁰-1+X,+Y,+Z

Table S2. Selected bond lengths and angles for compound **UOH-Ba2**.

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
U1-O16	2.139(7)	U2-O3	2.389(7)	U3-O5	2.317(7)
U1-O17	2.422(7)	U2-O13	2.450(7)	U3-O6	2.438(7)
U1-O18	1.838(7)	U2-O14	1.808(7)	U3-O10	2.196(7)
U1-O19	1.841(7)	U2-O15	1.794(7)	U3-O11	1.803(7)
U1-O20	2.213(7)	U2-O16	2.269(7)	U3-O12	1.828(7)
U1-O20 ¹	2.323(7)	U2-O16 ²	2.285(7)	U3-O13	2.321(7)
O18=U1=O19	175.9(3)	U2-O20 ¹	2.355(7)	U3-O17 ²	2.583(7)
		O15=U2=O14	176.7(3)	O11=U3=O12	178.7(3)
U4-O1	1.815(7)	U5-O4 ³	2.376(8)	U6-O4 ⁸	2.052(7)
U4-O2	1.784(7)	U5-O6	2.340(7)	U6-O4	2.052(7)
U4-O3	2.294(7)	U5-O7	1.798(8)	U6-O5	2.102(7)
U4-O4	2.404(7)	U5-O8	1.773(7)	U6-O5 ⁸	2.102(7)
U4-O5	2.393(7)	U5-O9	2.510(7)	U6-O6	2.056(7)
U4-O10 ⁵	2.241(7)	U5-O10	2.210(7)	U6-O6 ⁸	2.056(7)
U4-O17 ¹	2.454(7)	U5-O12 ⁷	2.522(7)		
O2=U4=O1	178.7(3)	O8=U5=O7	178.1(4)		
Ba-O1 ⁶	2.734(7)	Ba-O15 ¹	2.763(7)		
Ba-O2 ¹	2.717(7)	Ba-O18	2.747(8)		
Ba-O7 ⁴	2.891(8)	Ba-O19 ¹	2.947(8)		
Ba-O8 ⁹	2.754(8)	Ba-O20	3.073(7)		
Ba-O11 ⁴	2.778(7)	Ba-O21 ⁴	3.306(6)		

¹1-X,2-Y,1-Z; ²1/2-X,3/2-Y,1-Z; ³-1/2+X,-1/2+Y,+Z; ⁴1-X,1-Y,1-Z; ⁵1/2+X,1/2+Y,+Z; ⁶3/2-X,3/2-Y,1-Z; ⁷1/2-X,-1/2+Y,1/2-Z; ⁸1-X,+Y,1/2-Z; ⁹1/2+X,3/2-Y,1/2+Z

Table S3. Bond valence sums (BVS) for cations and anions in compound **UOH-Ba1**.

		U(1)	U(2)	Ba	Σ
Occ	*	1	1	0.5	
CN	#	7	7	11	
O1		1.71		0.12	2.10
				0.27	
O2		1.67		0.31	2.26
				0.27	
O3		0.70		0.07	2.01
		0.60			
		0.64			
O4			1.70	0.24	2.20
				0.26	
O5			1.64	0.12	1.76
O6			0.54	0.06	1.97
			0.72		
			0.65		
O7		0.54	0.59		1.13 (OH)
O8		0.73	0.74		1.47 (OH)
O9				0.21, 0.21	0.42 (H ₂ O)
Σ		6.60	6.59	2.14	

Table S4. Bond valence sums (BVS) for cations and anions in compound **UOH-Ba2**.

		U1	U2	U3	U4	U5	U6	Ba	Σ
Occ	*	1	1	1	1	1	0.5	1	
CN	#	6	7	7	7	7	6	11	
O1					1.57			0.30	1.87
O2					1.67			0.31	1.98
O3			0.52		0.63				1.15 (OH)
O4					0.51	0.53	1.00, 1.00		2.04
O5				0.60	0.52		0.91, 0.91		2.03
O6				0.47		0.57	0.99, 0.99		2.03
O7						1.63		0.19	1.82
O8						1.71		0.28	1.99
O9						0.41			0.41 (H ₂ O)
O10				0.76	0.69	0.74			2.19
O11				1.61				0.26	1.87
O12				1.53		0.40			1.94
O13			0.46	0.59					1.05 (OH)
O14			1.60						1.60
O15			1.64					0.27	1.91
O16	0.84		0.66, 0.64						2.14
O17	0.49			0.36	0.46				1.26 (OH)
O18	1.51							0.29	1.80
O19	1.50							0.17	1.67
O20	0.73, 0.59		0.56					0.12	2.00
O21								0.06, 0.06	0.06 (H ₂ O)
Σ		5.66	6.07	5.92	6.04	5.99	5.78	2.21	