

Electronic Supplementary Information

Filling the gaps of uranium oxide hydrates with magnesium(II) ions:
unique layered structures and the role of additional sodium(I) ions[†]

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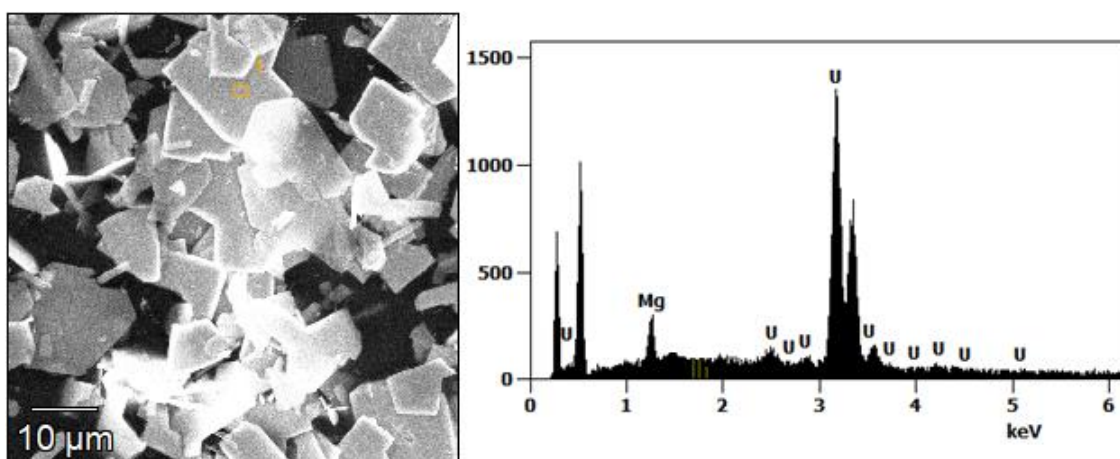


Fig. S1. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound **U-Mg1** with U:Mg = 3:1.

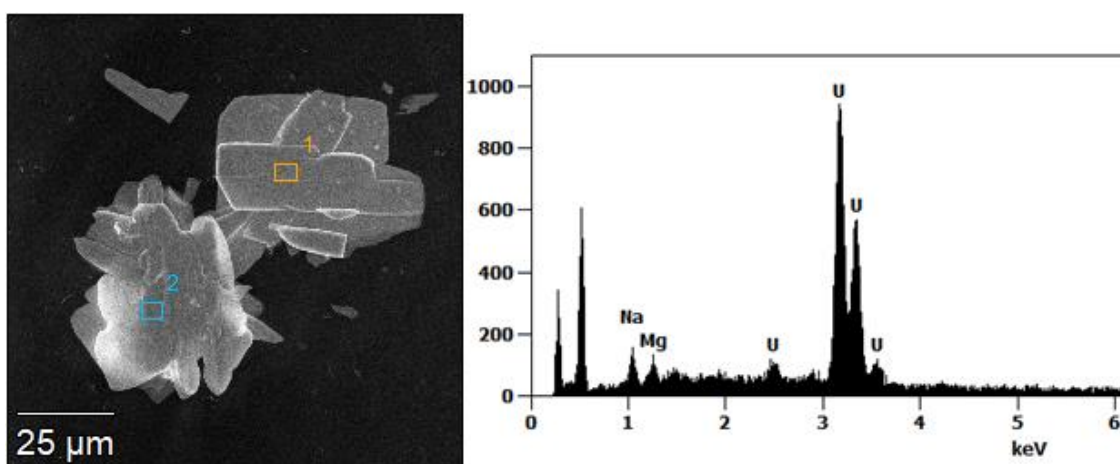


Fig. S2. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound **U-Mg2p** with U:Mg:Na = 6:1:2.

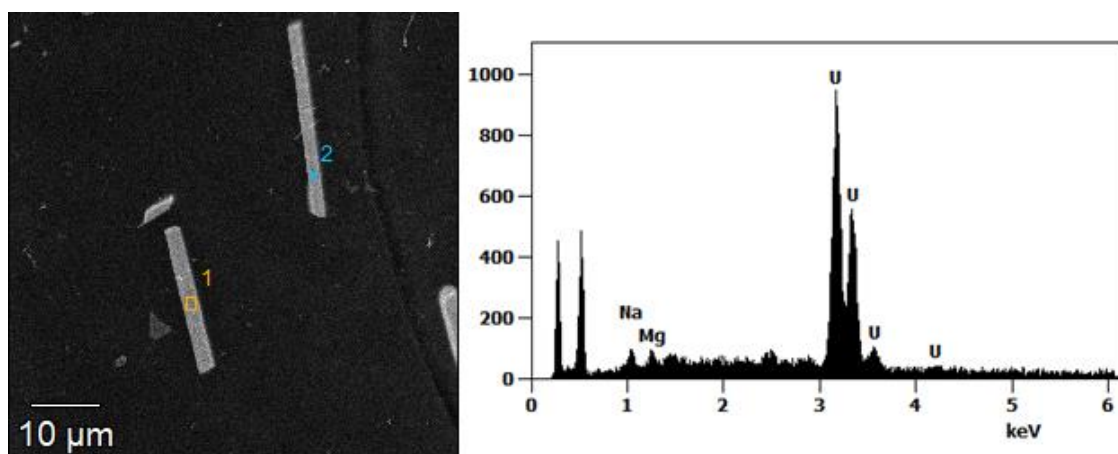


Fig. S3. A backscattered SEM image (top) and an EDS spectrum (bottom) of compound **U-Mg_{2n}** with U:Mg:Na = 8:1:2.

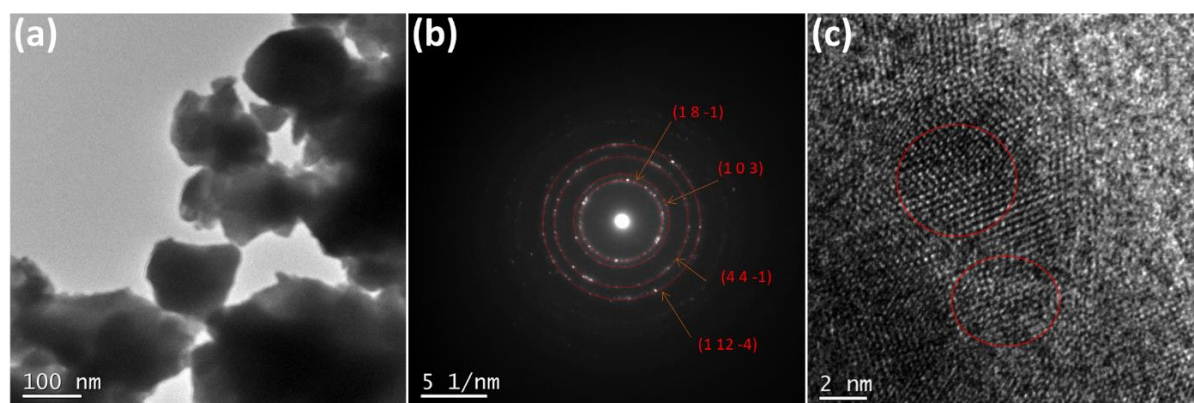


Fig. S4. TEM of **U-Mg₁**: a TEM bright field image of the crushed grains (a); a selected area electron diffraction (SAED) pattern showing crystalline diffractions and rings (b); and a HRTEM image showing lattice fringes with nano-domains in slightly different crystal orientations.

Table S1. Bond valence sums (BVS) for cations and anions in compound **U-Mg1**.

	U1	U2	U3	U4	U5	U6	Mg1	Mg2	Σ
Occ	*	1	1	1	1	1	1	1	
CN	#	6	7	7	6	7	7	6	6
O1	0.99								0.99
O2	0.93								0.93
O3	0.93	0.57				0.56			2.06
O4	0.99				0.53	0.55			2.07
O5	0.90	0.55	0.80						2.25
O6	0.85	0.63				0.55			2.03
O7		1.52							1.52
O8		1.57						0.40	1.97
O9			1.58				0.38		1.96
O10			1.50						1.50
O11		0.67	0.60	0.80					2.07
O12			0.34, 0.66	0.76					1.76
O13				1.19					1.19
O14				1.37					1.37
O15				0.82	0.64, 0.58				2.04
O16				0.80	0.57	0.68			2.05
O17					1.63				1.63
O18					1.53		0.38		1.91
O19			0.58		0.59				1.17 (OH)
O20		0.58				0.61			1.19 (OH)
O21						1.50		0.44	1.94
O22						1.61			1.61
O23							0.40		0.40 (H ₂ O)
O24							0.36		0.30 (H ₂ O)
O25							0.40		0.40 (H ₂ O)
O26							0.37		0.37 (H ₂ O)
O27								0.34	0.34 (H ₂ O)
O28								0.40	0.40 (H ₂ O)
O29								0.35	0.35 (H ₂ O)
O30								0.38	0.38 (H ₂ O)
Σ		5.59	6.09	5.84	5.75	6.07	6.05	2.29	2.31

Table S2. Bond valence sums (BVS) for cations and anions in compound **U-Mg2p**.

	U1	U2	U3	U4	U5	U6	Mg1	Na1	Na2	Σ
Occ	*	1	1	1	1	1	1	1	1	
CN	#	6	7	7	7	7	6	6	8	
O1	1.50							0.12, 0.19	0.18	1.99
O2	1.48									1.48
O3	0.70				0.48	0.69				1.97
O4	0.71	0.66	0.53							1.90
O5	0.69	0.71	0.70							2.10
O6		1.47						0.10, 0.21	0.12	1.90
O7		1.56								1.56
O8		0.58				0.58				1.16 (OH)
O9		0.26	0.52	0.55						1.43 (OH)
O10		0.71	0.70	0.74					0.08	2.15
O11				1.50					0.12, 0.16	1.78
O12				1.54						1.54
O13				0.76	0.75	0.71				2.22
O14			0.21	0.29	0.46					0.96 (OH)
O15					1.55				0.18	1.73
O16					1.56					1.56
O17				0.53	0.50	0.30				1.33 (OH)
O18			1.58					0.23	0.20	2.02
O19			1.60				0.40			2.00
O20						1.50	0.40			1.90
O21						1.50		0.17		1.67
O22	0.67				0.67	0.70			0.10	2.14
O23							0.31			0.31 (H ₂ O)
O24							0.38			0.38 (H ₂ O)
O25							0.36			0.36 (H ₂ O)
O26							0.35			0.35 (H ₂ O)
Σ	5.75	5.97	5.85	5.92	5.97	5.97	2.21	1.02	1.14	

Table S3. Bond valence sums (BVS) for cations and anions in compound **U-Mg2n**.

	U1	U2	U3	U4	Mg1	Na1	Σ
Occ	*	1	1	1	0.5	1	
CN	#	7	7	6	6	6	
O1	1.61						1.66
O2	1.65					0.19	1.84
O3	0.53, 0.34	0.44					1.31 (OH)
O4	0.60		0.48				1.08 (OH)
O5	0.80	0.66, 0.69					2.15
O6	0.37	0.41		0.44			1.22 (OH)
O7		1.57				0.15	1.72
O8		1.62				0.10	1.72
O9		0.60	0.58				1.18 (OH)
O10			1.56			0.18	1.74
O11			1.56		0.37, 0.37		1.93
O12			0.40, 0.64	0.80			1.84
O13				1.44		0.05	1.49
O14				1.58		0.17	1.75
O15			0.62	0.74, 0.62			1.98
O16					0.38, 0.38		0.76 (H ₂ O)
O17					0.32, 0.32		0.64 (H ₂ O)
Σ	5.90	6.00	5.86	5.62	2.13	0.85	