

Supporting Information

Framework uranyl silicates: crystal chemistry and a new route for the synthesis.

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Table S1. The types of Si_nO_m complexes in the structures of uranyl silicate.

Complex type	Dimensions	Chemical formula	Space group	$a, \text{Å} / \alpha, ^\circ$	$b, \text{Å} / \beta, ^\circ$	$c, \text{Å} / \gamma, ^\circ$	Ref.
$[\text{SiO}_4]^{4-}$	0D	α -uranophane, $\text{Ca}[(\text{UO}_2)_2(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_5$	$P2_1$	15.909	7.002 / 97.27	6.665	[1]
		β -uranophane, $\text{Ca}[(\text{UO}_2)_2(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_5$	$P2_1/a$	13.966	15.443 / 91.38	6.632	[1]
		kasolite, $\text{Pb}[(\text{UO}_2)(\text{SiO}_4)](\text{H}_2\text{O})$	$P2_1/c$	6.70	6.93 / 105.00	13.28	[2]
		boltwoodite, $\text{K}[(\text{UO}_2)(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_{1.5}$	$P2_1/m$	7.077	7.060 / 104.98	6.648	[3]
		natroboltwoodite, $\text{Na}[(\text{UO}_2)(\text{SiO}_3\text{OH})](\text{H}_2\text{O})$	$P2_12_12_1$	27.40	7.02	6.65	[3]
		sklodowskite, $\text{Mg}[(\text{UO}_2)(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_6$	$C2/m$	17.382	7.047 / 105.88	6.610	[2]
		cuprosklodowskite, $\text{Cu}[(\text{UO}_2)(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_6$	$P-1$	7.052 / 109.23	9.267 / 89.84	6.655 / 110.01	[2]
		oursinite, $\text{Co}[(\text{UO}_2)(\text{SiO}_3\text{OH})]_2(\text{H}_2\text{O})_6$	$Cmca$	7.04	17.55	12.73	[4]
		swamboite, $\text{Nd}_{0.333}[(\text{UO}_2)(\text{SiO}_3\text{OH})](\text{H}_2\text{O})_{2.41}$	$P2_1/a$	6.65	6.98 / 102.59	8.80	[5]
		soddyite, $[(\text{UO}_2)_2(\text{SiO}_4)](\text{H}_2\text{O})_2$	$Fddd$	8.334	11.212	18.668	[6]
$\text{Na}_2[(\text{UO}_2)_2(\text{SiO}_4)\text{F}_2]$	$I4_1/amd$	6.975	6.975	18.31	[7]		
$[\text{Si}_2\text{O}_7]^{6-}$	0D	$\text{Na}_6[(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$C2/m$	22.323	7.463 / 99.377	5.777	[8]
		$[\text{Na}_3\text{K}_3][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2](\text{H}_2\text{O})_2$	$P-1$	5.798 / 103.59	7.588 / 102.88	12.807 / 90.06	[9]
		$[\text{Na}_3\text{Rb}_3][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$P-1$	5.799 / 78.265	7.575 / 79.14	12.937 / 89.94	[9]
		$[\text{K}_3\text{Cs}_4\text{F}][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$Cmc2_1$	7.809	22.282	14.086	[10]
		$[\text{NaRb}_6\text{F}][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$Pnnm$	11.143	13.515	7.887	[10]
		$[\text{NaK}_6\text{F}][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$Pnnm$	11.082	13.115	7.842	[11]
		$[\text{KK}_6\text{Cl}][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$Pnnm$	11.083	13.585	7.869	[11]
		$[\text{Cs}_2\text{Cs}_5\text{F}][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2]$	$P2_1/n$	7.556	9.864 / 91.71	18.554	[11]
		$[\text{K}_2\text{Ca}_4][(\text{UO}_2)(\text{Si}_2\text{O}_7)_2]$	$P-1$	6.635 / 98.32	6.679 / 93.62	9.699 / 112.31	[12]
		$[\text{K}_4\text{Na}_2][(\text{UO}_2)_3(\text{Si}_2\text{O}_7)_2](\text{H}_2\text{O})_3$	$C2/m$	13.154	15.159 / 104.64	5.824	[12]
$[\text{K}_8\text{K}_5\text{F}][\text{U}_6(\text{Si}_8\text{O}_{40})]$	$P2_12_12_1$	11.803	13.698	26.679	[11]		
$[\text{Si}_4\text{O}_{12}]^{8-}$	0D	$\text{Cs}_2[\text{USiO}_6]$	$Immm$	8.581	13.001	13.881	[13]
		$\text{Rb}_2[\text{USiO}_6]$	$Immm$	8.492	12.668	13.508	[13]

		Rb ₂ [(UO ₂)(Si ₂ O ₆)](H ₂ O)	<i>P2₁/n</i>	7.699	20.97 / 97.92	12.05	[14]
		[RbNa][[(UO ₂)(Si ₂ O ₆)](H ₂ O)]	<i>P-1</i>	7.367 / 78.02	7.869 / 75.01	8.177 / 83.74	[15]
		[K ₄ Ca][U(Si ₂ O ₇) ₂]	<i>P-1</i>	6.635 / 98.32	6.679 / 93.62	9.699 / 112.31	[12]
		[K ₈ K ₅ F][U ₆ (Si ₈ O ₄₀)]	<i>P2₁2₁2₁</i>	11.803	13.698	26.679	[16]
[Si ₄ O ₁₂] ⁸⁻	0D	[Cs ₉ Cs ₆ Cl][[(UO ₂) ₇ (Si ₆ O ₁₇) ₂ (Si ₄ O ₁₂)]	<i>P-1</i>	7.350 / 89.29	15.332 / 89.99	17.255 / 76.45	[16]
[Si ₄ O ₁₂ (OH)] ⁹⁻	0D	K ₅ [(UO ₂) ₂ (Si ₄ O ₁₂ (OH))]	<i>Pbcm</i>	13.127	12.264	22.233	[17]
[Si ₂ O ₆] ⁴⁻	1D	K ₂ [(UO ₂)Si ₂ O ₆]	<i>C2/c</i>	21.695	14.416 / 136.71	16.073	[18]
		Ba[(UO ₂)(Si ₂ O ₆)]	<i>Cmcm</i>	5.703	16.464	7.496	[19]
		α-Cs ₂ [(UO ₂)(Si ₂ O ₆)]	<i>lbca</i>	15.137	15.295	16.401	[17]
		Rb ₂ [(UO ₂)(Si ₂ O ₆)](H ₂ O) _{0.5}	<i>Pbca</i>	14.627	15.145	16.645	[14]
		Cs ₂ [(UO ₂)(Si ₂ O ₆)](H ₂ O) _{0.5}	<i>Pbca</i>	15.047	15.427	16.732	[14]
		Rb ₂ [(UO ₂)Si ₂ O ₆]	<i>I2/a</i>	14.993	14.803 / 90.72	16.238	[20]
		β-Cs ₂ [(UO ₂)(Si ₂ O ₆)]	<i>C2</i>	12.136	10.043 / 95.85	7.789	[20]
[Si ₄ O ₁₀] ⁴⁻	1D	[Cs ₃ F][[(UO ₂)(Si ₄ O ₁₀)]	<i>Imma</i>	15.476	7.815	12.756	[16]
		Na ₂ [(UO ₂)(Si ₄ O ₁₀)](H ₂ O) _{0.5}	<i>I4/mcm</i>	18.033	18.033	7.775	[21]
		Cs ₂ [(UO ₂)(Si ₄ O ₁₀)]	<i>Cmca</i>	7.72	19.87	24.02	[21]
		Rb ₂ [(UO ₂)(Si ₄ O ₁₀)]	<i>P2₁/c</i>	10.348	23.730 / 110.48	7.628	[21]
[Si ₅ O ₁₃] ⁶⁻	1D	haiweeite, Ca[(UO ₂) ₂ (Si ₅ O ₁₂)(OH) ₂](H ₂ O) ₆	<i>Pbcn</i>	18.30	14.23	17.92	[22]
[Si ₆ O ₁₇] ¹⁰⁻	1D	[Cs ₂ Cs ₅ F][[(UO ₂) ₂ (Si ₆ O ₁₇)]	<i>P2₁2₁2</i>	10.374	19.280	7.180	[11]
		[Cs ₉ Cs ₆ Cl][[(UO ₂) ₇ (Si ₆ O ₁₇) ₂ (Si ₄ O ₁₂)]	<i>P-1</i>	7.350 / 89.29	15.332 / 89.99	17.255 / 76.45	[11]
[Si ₈ O ₂₀] ⁸⁻	1D	Rb ₄ [(UO ₂)(Si ₈ O ₂₀)]	<i>P-1</i>	6.844 / 72.79	8.314 / 88.74	11.273 / 77.95	[14]
[Si ₁₀ O ₃₀] ²⁰⁻	1D	K ₁₄ [(UO ₂) ₃ Si ₁₀ O ₃₀]	<i>P2₁/c</i>	12.902	11.125 / 90.80	13.952	[21]
[Si ₄ O ₁₀] ⁴⁻	2D	KNa ₃ [(UO ₂) ₂ (Si ₄ O ₁₀) ₂](H ₂ O) ₄	<i>C2</i>	12.782	13.654 / 119.24	8.268	[23]
		α-K ₂ [(UO ₂)Si ₄ O ₁₀]	<i>P2₁/n</i>	8.297	7.7403 / 104.83	8.7906	[21]
		β-K ₂ [(UO ₂)(Si ₄ O ₁₀)]	<i>C2/c</i>	17.933	6.793 / 125.52	11.744	[8]
		Na ₄ [(UO ₂)(Si ₄ O ₁₀) ₂](H ₂ O) ₄	<i>P2₁/n</i>	7.048	11.413 / 90.56	12.027	[2]

		$\text{Na}_2[(\text{UO}_2)(\text{Si}_4\text{O}_{10})](\text{H}_2\text{O})_{2.1}$	<i>C2/m</i>	12.772	13.614 / 119.26	8.247	[15]
$[\text{Si}_5\text{O}_{13}]^{6-}$	2D	wecksite, $\text{K}_2[(\text{UO}_2)_2(\text{Si}_5\text{O}_{13})](\text{H}_2\text{O})_4$	<i>C2/m</i>	14.19	14.22	9.63	[2]
		$\text{Rb}_2[(\text{UO}_2)\text{Si}_5\text{O}_{13}]$	<i>C222</i>	7.118	17.949	7.057	[24]
$[\text{Si}_8\text{O}_{20}]^{8-}$	2D	$\text{K}_4[(\text{UO}_2)_2(\text{Si}_8\text{O}_{20})](\text{H}_2\text{O})_4$	<i>P2_1</i>	14.179	14.179	14.965	[8]
$[\text{Si}_{10}\text{O}_{22}]^{4-}$	2D	$\text{Cs}_2[(\text{UO}_2)\text{Si}_{10}\text{O}_{22}]$	<i>P2_1/c</i>	12.251	8.052 / 90.01	23.380	[25]

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Table. S2. Selected interatomic bonds in the structure of Rb₂[(UO₂)₂(Si₈O₁₉)](H₂O)_{2.5}

U1-O2	1.796(4)	Rb1-OW2	2.582(14)	Rb2-OW1	2.907(6)
U1-O1	1.816(4)	Rb1-O1	2.943(3)	Rb2-O1	2.973(3)
<U-O _{ap} >	1.806	Rb1-O1	2.943(3)	Rb2-O1	2.973(3)
U1-O3	2.217(4)	Rb1-OW1	3.056(6)	Rb2-O10	3.001(4)
U1-O4	2.326(3)	Rb1-O5	3.115(3)	Rb2-OW3	3.059(8)
U1-O6	2.328(3)	Rb1-O5	3.115(3)	Rb2-OW3	3.059(8)
U1-O4	2.481(3)	Rb1-O9	3.125(4)	Rb2-O13	3.089(3)
U1-O6	2.499(3)	Rb1-O2	3.257(3)	Rb2-O13	3.089(3)
<U-O _{eq} >	2.370	Rb1-O2	3.257(3)	Rb2-O4	3.477(3)
Si1-O8	1.5946(10)	Rb1-OW3	3.376(8)	Rb2-O4	3.477(3)
Si1-O3	1.595(4)	Rb1-OW3	3.376(8)	Rb2-O1	3.548(3)
Si1-O11	1.619(3)	Rb1-O6	3.660(3)	Rb2-O1	3.548(3)
Si1-O12	1.626(3)	<Rb1-O>	2.908	<Rb2-O>	2.938
<Si1-O>	1.609				
Si2-O6	1.611(3)	Si3-O12	1.588(3)	Si4-O11	1.588(3)
Si2-O4	1.614(3)	Si3-O13	1.609(3)	Si4-O9	1.6075(13)
Si2-O5	1.620(3)	Si3-O10	1.6130(13)	Si4-O7	1.608(3)
Si2-O13	1.623(3)	Si3-O7	1.614(3)	Si4-O5	1.610(3)
<Si2-O>	1.617	<Si3-O>	1.606	<Si4-O>	1.603

Table. S3. Selected interatomic bonds in the structure of (K,Rb)₂[(UO₂)(Si₁₀O₂₂)]

U1-O1	1.798(4)	U2-O2	1.794(4)	Si1-O11	1.587(3)
U1-O1	1.798(4)	U2-O2	1.794(4)	Si1-O13	1.610(3)
<U1-O _{ap} >	1.798	<U2-O _{ap} >	1.794	Si1-O7	1.610(3)
U1-O6	2.275(3)	U2-O5	2.269(3)	Si1-O4	1.6124(17)
U1-O6	2.275(3)	U2-O5	2.269(3)	<Si1-O>	1.605
U1-O6	2.275(3)	U2-O5	2.269(3)	Si2-O6	1.582(3)
U1-O6	2.275(3)	U2-O5	2.269(3)	Si2-O14	1.605(4)
<U1-O _{eq} >	2.275	<U2-O _{eq} >	2.269	Si2-O7	1.624(3)
				Si2-O10	1.6269(1)
Rb1-O8	3.015(4)	K2-O5	2.877(3)	<Si2-O>	1.609
Rb1-O1	3.064(5)	K2-O5	2.877(4)	Si3-O14	1.577(4)
Rb1-O6	3.114(3)	K2-O10	2.921(5)	Si3-O13	1.578(3)
Rb1-O6	3.114(3)	K2-O2	3.042(5)	Si3-O16	1.582(4)
Rb1-O9	3.410(3)	K2-O7	3.104(4)	Si3-O12	1.6025(1)
Rb1-O9	3.410(3)	K2-O7	3.104(4)	<Si3-O>	1.585
Rb1-O5	3.443(3)	K2-O6	3.294(3)	Si4-O16	1.573(4)
Rb1-O5	3.443(3)	K2-O6	3.294(3)	Si4-O15	1.5740(1)
Rb1-O12	3.484(5)	K2-O10	3.403(5)	Si4-O3	1.5905(16)
Rb1-O8	3.510(5)	<K1-O>	3.102	Si4-O9	1.604(3)
<Rb1-O>	3.301			<Si4-O>	1.585
Si5-O5	1.586(3)				
Si5-O11	1.610(3)				
Si5-9	1.619(3)				
Si5-O8	1.6271(19)				
<Si5-O>	1.611				

Table. S4. Selected interatomic bonds in the structure of [Rb₃Cl][(UO₂)(Si₄O₁₀)]

U1-O3	1.801(5)	Rb1-O1	3.053(4)	Rb2-O3	2.862(5)
U1-O3	1.801(5)	Rb1-O1	3.053(4)	Rb2-O5	3.244(6)
<U1-O _{ap} >	1.801	Rb1-O1	3.053(4)	Rb2-O5	3.244(5)
U1-O1	2.252(4)	Rb1-O1	3.053(4)	Rb2-O1	3.306(4)
U1-O1	2.252(4)	Rb1-Cl1	3.1256(7)	Rb2-O1	3.306(4)
U1-O1	2.252(4)	Rb1-Cl1	3.1256(7)	Rb2-Cl1	3.4010(8)
U1-O1	2.252(4)	Rb1-O2	3.164(6)	Rb2-Cl1	3.4010(8)
<U1-O _{eq} >	2.252	Rb1-O2	3.164(6)	<Rb2-O>	3.252
Si1-O1	1.583(4)	Rb1-O3	3.300(5)		
Si1-O4	1.6085(19)	Rb1-O3	3.300(5)		
Si1-O2	1.6085(15)	<Rb1-O>	3.139		
Si1-O5	1.620(2)				
<Si1-O>	1.605				

Table. S5. Selected interatomic bonds in the structure of [Cs₃Cl][(UO₂)(Si₄O₁₀)]

U1-O3	1.805(4)	RB1-O2	3.146(5)	RB2-O3	3.089(4)
U1-O3	1.805(4)	RB1-O2	3.146(5)	RB2-O5	3.355(5)
<U1-O _{ap} >	1.805	RB1-O1	3.192(3)	RB2-O1	3.433(3)
U1-O1	2.261(3)	RB1-O1	3.192(3)	RB2-O1	3.433(3)
U1-O1	2.261(3)	RB1-O1	3.192(3)	RB2-O5	3.476(5)
U1-O1	2.261(3)	RB1-O1	3.192(3)	RB2-Cl1	3.5025(4)
U1-O1	2.261(3)	RB1-O3	3.209(4)	RB2-Cl1	3.5025(4)
<U1-O _{eq} >	2.261	RB1-O3	3.209(4)	<Rb2-O>	3.398
Si1-O1	1.585(3)	RB1-Cl1	3.2120(5)		
Si1-O5	1.6139(15)	RB1-Cl1	3.2120(5)		
Si1-O2	1.6190(14)	<Rb1-O>	3.190		
Si1-O4	1.6191(17)				
<Si1-O>	1.609				

Figure S1. Powder XRD of $(\text{K,Rb})_2[(\text{UO}_2)(\text{Si}_{10}\text{O}_{22})]$ and $\text{Rb}_2[(\text{UO}_2)_2(\text{Si}_8\text{O}_{19})](\text{H}_2\text{O})_{2.5}$

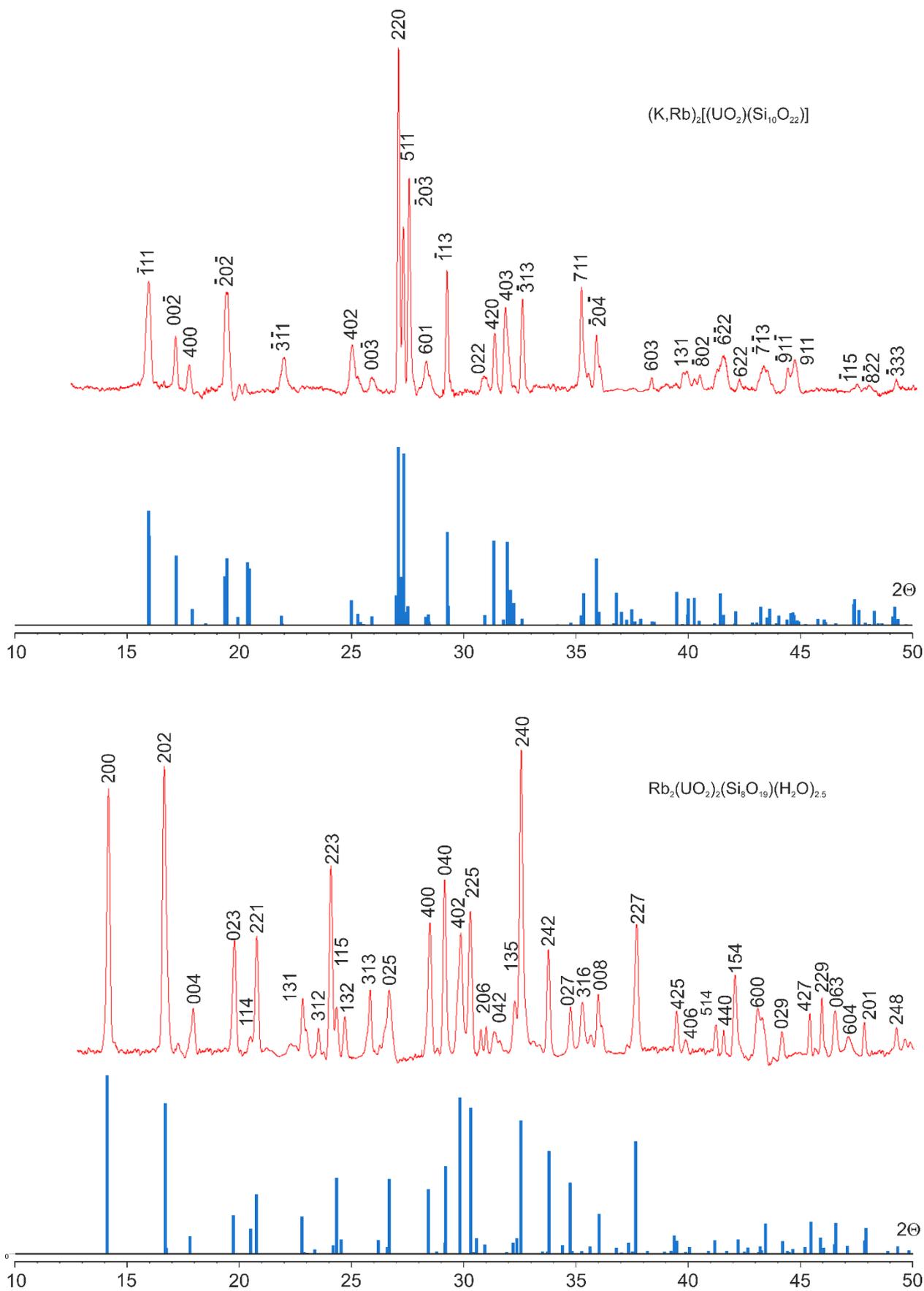


Figure S2. Powder XRD of $[\text{Rb}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$ and $[\text{Cs}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$

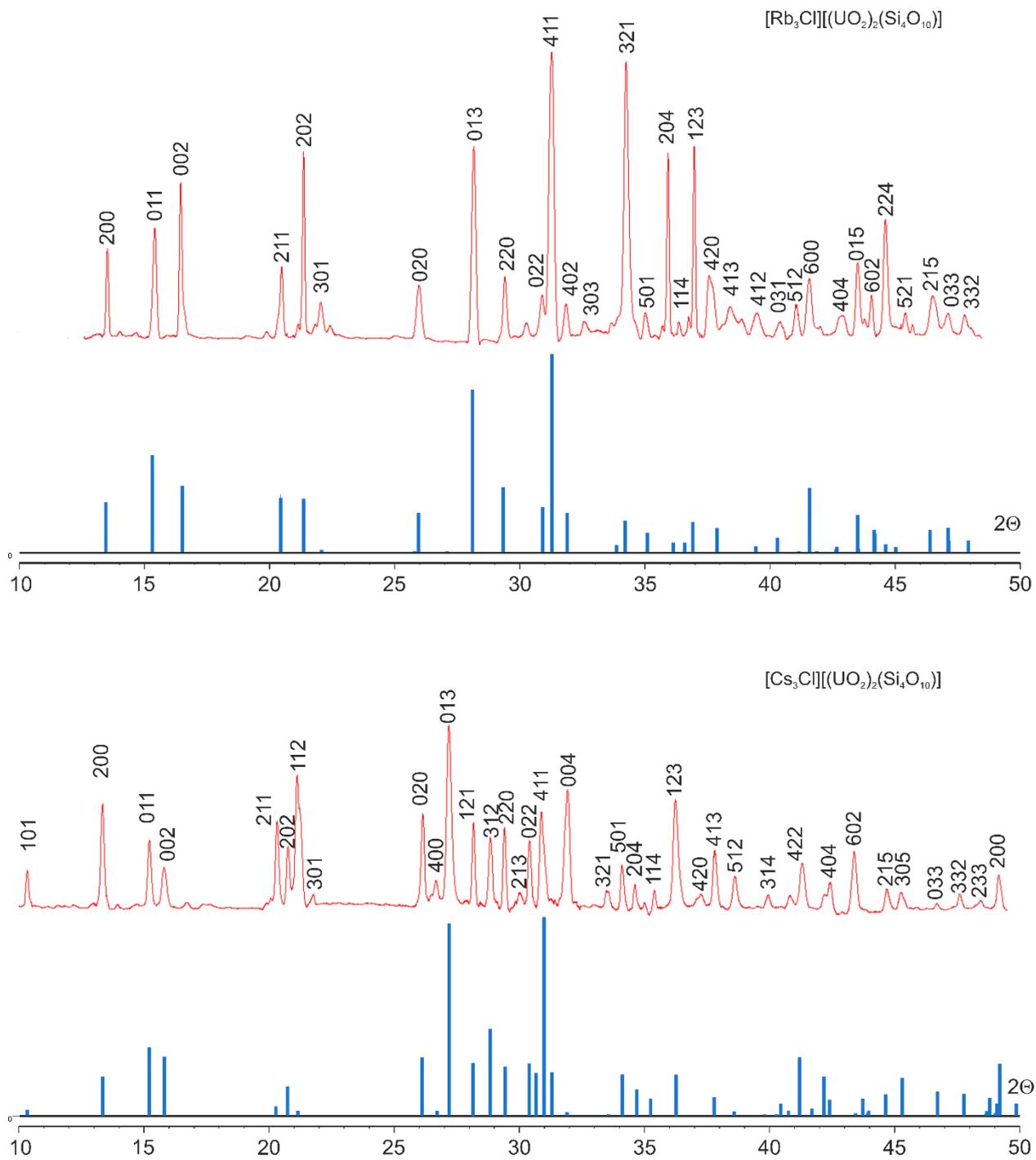


Figure S3. IR absorption spectra of $\text{Rb}_2[(\text{UO}_2)_2(\text{Si}_8\text{O}_{19})](\text{H}_2\text{O})_{2.5}$ (a), $(\text{K,Rb})_2[(\text{UO}_2)(\text{Si}_{10}\text{O}_{22})]$ (b), $[\text{Rb}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$ (c) and $[\text{Cs}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$ (d)

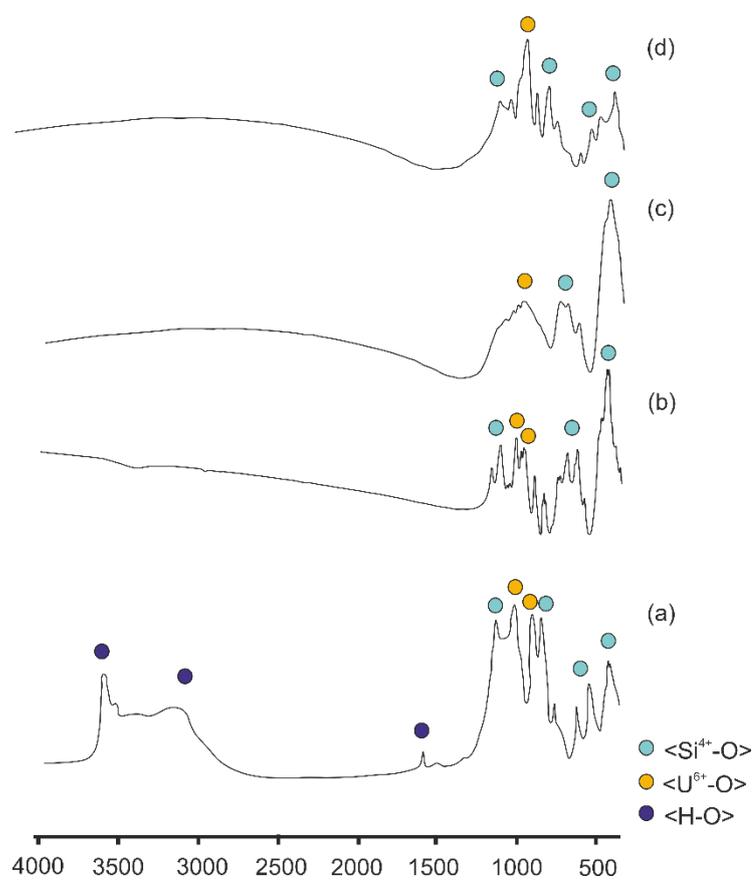


Table S6. Chemical data (in wt %) for $(\text{K,Rb})_2[(\text{UO}_2)(\text{Si}_{10}\text{O}_{22})]$

Constituent	Mean	Range	Stand. Dev. (2σ)	Reference Material
UO ₃	28,67	28.57 – 28.77	0.10	UO ₂
SiO ₂	61,93	61.80 – 62.06	0.13	CaSiO ₃
Rb ₂ O	3,29	3.10 – 3.48	0.19	RbCl
K ₂ O	7.98	7.79 – 8.17	0.19	KCl
Total	101.87			

The empirical formula calculated on the basis of 24 oxygen atoms per formula unit is $(\text{Rb}_{1,65}\text{K}_{0,34})[(\text{U}_{0,98}\text{O}_2)(\text{Si}_{10,04}\text{O}_{22})]$

Table S7. Chemical data (in wt %) for $\text{Rb}_2[(\text{UO}_2)_2(\text{Si}_8\text{O}_{19})](\text{H}_2\text{O})_{2.5}$

Constituent	Mean	Range	Stand. Dev. (2σ)	Reference Material
UO ₃	44,48	44.38 – 44.58	0.10	UO ₂
SiO ₂	37,37	37.24 – 37.50	0.13	CaSiO ₃
Rb ₂ O	14,61	14.42 – 14.80	0.19	RbCl
H ₂ O	3,52	3.33 – 3.71	0.19	
Total	99.99			

The empirical formula calculated on the basis of 25.5 oxygen atoms per formula unit is $\text{Rb}_{2,01}[(\text{UO}_2)_2(\text{Si}_{7,99}\text{O}_{19})](\text{H}_2\text{O})_{2,52}$

Table S8. Chemical data (in wt %) for $[\text{Rb}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$

Constituent	Mean	Range	Stand. Dev. (2σ)	Reference Material
UO ₃	33,58	33.48 – 33.68	0.10	UO ₂
SiO ₂	28,58	28.45 – 28.71	0.13	CaSiO ₃
Rb ₂ O	33,59	33.40 – 33.68	0.19	RbCl
Cl	4,25	4.06 – 4.44	0.19	Pyromorphite
O=Cl ₂	0,96			
Total	99.03			

The empirical formula calculated on the basis of 13 oxygen atoms per formula unit is $[\text{Rb}_{3,02}\text{Cl}][(\text{U}_{0,99}\text{O}_2)(\text{Si}_{4,01}\text{O}_{10})]$

Table S9. Chemical data (in wt %) for $[\text{Cs}_3\text{Cl}][(\text{UO}_2)(\text{Si}_4\text{O}_{10})]$

Constituent	Mean	Range	Stand. Dev. (2σ)	Reference Material
UO_3	29,29	29.19 – 29.39	0.10	UO_2
SiO_2	24,25	24.12 – 24.38	0.13	CaSiO_3
Cs_2O	42,86	42.69 – 43.03	0.17	CsCl
Cl	3,60	3.41 – 3.79	0.19	Pyromorphite
$\text{O}=\text{Cl}_2$	0,814			
Total	99.19			

The empirical formula calculated on the basis of 13 oxygen atoms per formula unit is $[\text{Cs}_3\text{Cl}][(\text{U}_{1.01}\text{O}_2)(\text{Si}_{3.98}\text{O}_{10})]$

Table S10. Bond valences parameters in structure of $\text{Rb}_2[(\text{UO}_2)_2(\text{Si}_8\text{O}_{19})](\text{H}_2\text{O})_{2.5}$.

	U1	Si1	Si2	Si3	Si4	Rb1	Rb2	Σ
O1	1.63					0.13×2	0.13×2 0.04×2	1.93
O2	1.70					0.07×2		1.77
O3	0.70	1.08						1.78
O4	0.55 0.4		1.03				0.04×2	2.02
O5			1.01		1.04	0.09×2		2.14
O6	0.55 0.38		1.03			0.03		1.99
O7				1.03	1.04			2.07
O8		1.08						2.16
O9					1.04	0.09		2.17
O10				1.03			0.12	2.18
O11		1.01			1.10			2.11
O12		0.99		1.10				2.13
O13			1.00	1.04			0.1×2	2.14
OW1						0.12	0.14	0.26
OW2*¹						0.34		0.34
OW3*²						0.05	0.11×2	0.16
Σ	5.91	4.16	4.07	4.19	4.22	1.28	1.08	

*¹ OW1 = 0.40, *² OW1 = 0.55

Table. S11. The BVS parameters for Si-O bonds in the structure of uranyl silicates.

compound	site	<Si-O>	S _{ij}
K ₁₄ (UO ₂) ₃ Si ₁₀ O ₃₀	Si1	1.628	3.97
	Si2	1.620	4.05
	Si3	1.625	3.99
	Si4	1.628	3.97
	Si5	1.632	3.93
K ₂ (UO ₂)Si ₂ O ₆	Si1	1.604	4.22
	Si2	1.611	4.14
	Si3	1.614	4.10
	Si4	1.612	4.13
β-K ₂ (UO ₂)Si ₄ O ₁₀	Si1	1.588	4.39
	Si2	1.596	4.31
K ₄ (UO ₂) ₂ Si ₈ O ₂₀ (H ₂ O) ₄	Si1	1.628	4.04
	Si2	1.577	4.53
	Si3	1.594	4.37
	Si4	1.653	3.75
	Si5	1.612	4.19
	Si6	1.591	4.36
	Si7	1.641	3.90
	Si8	1.610	4.17
	Si9	1.579	4.49
	Si10	1.629	4.01
	Si11	1.634	3.91
	Si12	1.624	4.03
	Si13	1.603	4.27
	Si14	1.612	4.15
	Si15	1.633	3.98
	Si16	1.599	4.33
Na ₆ (UO ₂) ₃ (Si ₂ O ₇) ₂	Si1	1.602	4.24
Na ₂ UO ₂ SiO ₄	Si1	1.603	4.22
Ba(UO ₂)(Si ₂ O ₆)	Si1	1.596	4.31
Na ₂ (UO ₂) ₂ (SiO ₄)F ₂	Si1	1.607	4.18
Na ₂ (UO ₂)(Si ₄ O ₁₀)(H ₂ O) _{2.1}	Si1	1.613	4.12
	Si2	1.611	4.12
Cs ₂ (UO ₂)Si ₂ O ₆	Si1	1.616	4.08
	Si2	1.627	3.97
K(UO)Si ₂ O ₆	Si1	1.619	4.05
	Si2	1.619	4.05
	Si3	1.623	4.01
	Si4	1.619	4.05
Rb ₂ (UO ₂)(Si ₂ O ₆)(H ₂ O)	Si1	1.633	3.91
	Si2	1.631	3.93
	Si3	1.642	3.82
	Si4	1.636	3.88
Cs ₂ (UO ₂)(Si ₂ O ₆)(H ₂ O) _{0.5}	Si1	1.626	3.98
	Si2	1.629	3.95
	Si3	1.625	3.99
	Si4	1.621	4.03
Rb ₄ (UO ₂) ₂ (Si ₈ O ₂₀)	Si1	1.619	4.06

	Si2	1.622	4.02
	Si3	1.629	3.95
	Si4	1.605	4.20
Rb ₂ (UO ₂)Si ₂ O ₆	Si1	1.618	4.07
	Si2	1.613	4.12
	Si3	1.613	4.12
	Si4	1.617	4.08
Cs ₂ (UO ₂)Si ₂ O ₆	Si1	1.621	4.03
	Si2	1.606	4.19
(K ₃ Cs ₄ F)((UO ₂) ₃ (Si ₂ O ₇) ₂)	Si1	1.620	4.06
	Si2	1.634	3.90
(NaRb ₆ F)((UO ₂) ₃ (Si ₂ O ₇) ₂)	Si1	1.628	3.97
K ₂ Ca ₄ ((UO ₂)(Si ₂ O ₇) ₂)	Si1	1.619	4.05
	Si2	1.616	4.08
Cs ₂ UO ₂ Si ₁₀ O ₂₂	Si1	1.596	4.30
	Si2	1.614	4.10
	Si3	1.597	4.29
	Si4	1.598	4.28
	Si5	1.603	4.23
	Si6	1.604	4.21
	Si7	1.604	4.21
	Si8	1.611	4.15
	Si9	1.598	4.28
	Si10	1.598	4.28
Cs ₂ UO ₂ SiO ₄	Si1	1.615	4.10
K ₄ CaU(Si ₂ O ₇) ₂	Si1	1.595	4.32
K(UO)Si ₂ O ₆	Si1	1.611	4.14
K ₈ (K ₅ F)U ₆ Si ₈ O ₄₀	Si1	1.612	4.13
	Si2	1.618	4.06
	Si3	1.616	4.09
	Si4	1.612	4.13
	Si5	1.634	3.90
	Si6	1.629	3.95
	Si7	1.620	4.04
	Si8	1.633	3.92

Table S12. Bond valences parameters in structure of (K,Rb)₂[(UO₂)(Si₁₀O₂₂)]

	O1	O2	O3	O4	O5	O6	O7	O8	O9	O10	O11	O12	O13	O14	O15	O16	Σ
U1	1.69×2					0.62×4											5.84
U2		1.70×2			0.62×4												5.90
Si1				1.03			1.04				1.10		1.04				4.20
Si2						1.11	1.00			0.99				1.05			4.16
Si3												1.06	1.13	1.13		1.11	4.43
Si4			1.09						1.05						1.14	1.14	4.42
Si5					1.10			0.99	1.01		1.04						4.14
Rb1¹	0.10					0.09×2		0.12 0.04	0.05×2			0.04					0.68
K1		0.11			0.15×2	0.06×2	0.1×2			0.14 +0.05							0.93
Σ	1.79	1.81	2.18	2.06	1.87	1.88	2.14	2.1	2.11	2.12	2.14	2.16	2.17	2.18	2.28	2.25	

Note. ¹ The calculation was carried out using parameters for Rb-O bond. Rb1/K1 = 0.366(7)/0.634(7).

Table S13. Bond valences parameters in structure of [Rb₃Cl][(UO₂)(Si₄O₁₀)]

	O1	O2	O3	O4	O5	Cl1	
U1	0.65×4		1.68×2				5.94
Si1	1.11	1.04		1.04	1.01		4.20
Rb1	0.11×4	0.08×2	0.06×2			0.2×2	1.12
Rb2	0.06×2		0.16		0.07×2	0.12×2	0.66
	1.93	2.16	1.9	2.08	2.09	0.64	

Table S14. Bond valences parameters in structure of [Cs₃Cl][(UO₂)(Si₄O₁₀)]

	O1	O2	O3	O4	O5	F1	F2	F3	
U1		1.63×2	0.66×4						5.90
Si1	1.02		1.11	1.02	1.03				4.18
Rb1		0.08×2				0.12×2	0.1×2	0.07×2 0.08×2	0.89
Rb2		0.18	0.08×2		0.05×2	0.05×2			0.57
	2.04	1.89	1.77	2.04	2.11	0.34	0.2	0.3	