Electronic Supplementary Material (ESI) for CrystEngComm.

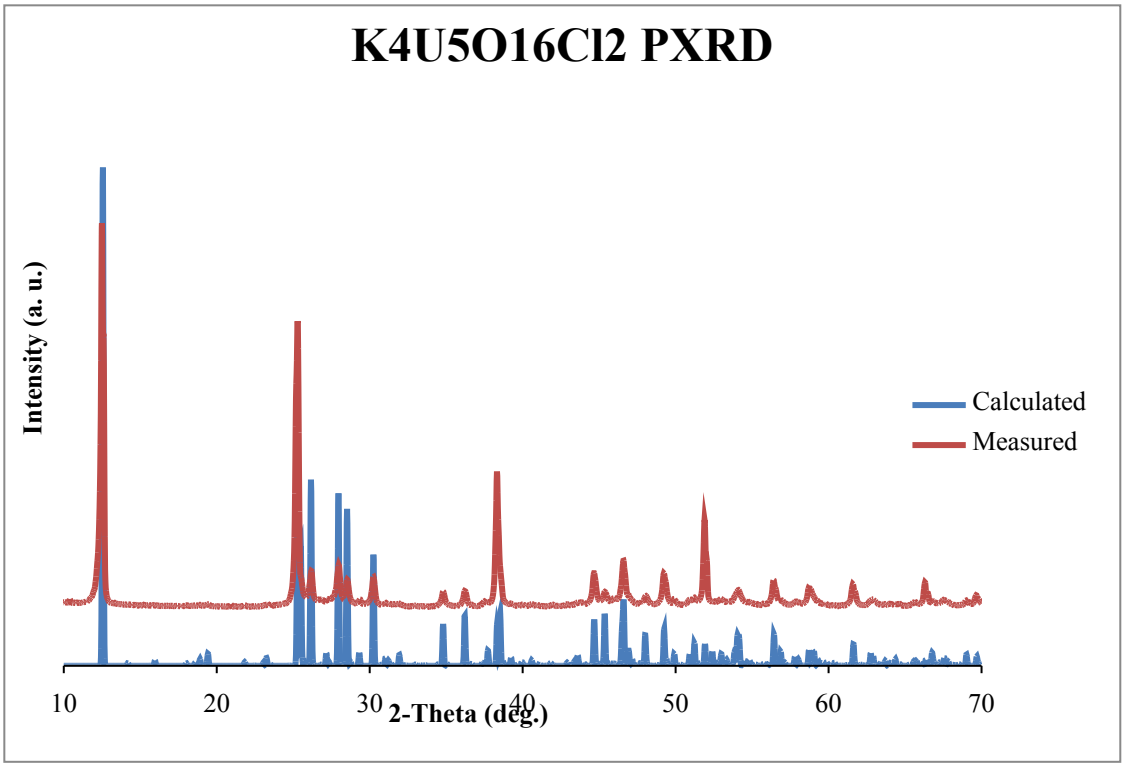

Figure S1: Normalized PXRD patterns for $K_4U_5O_{16}Cl_2$, with the measured pattern in red and the calculated pattern in blue.

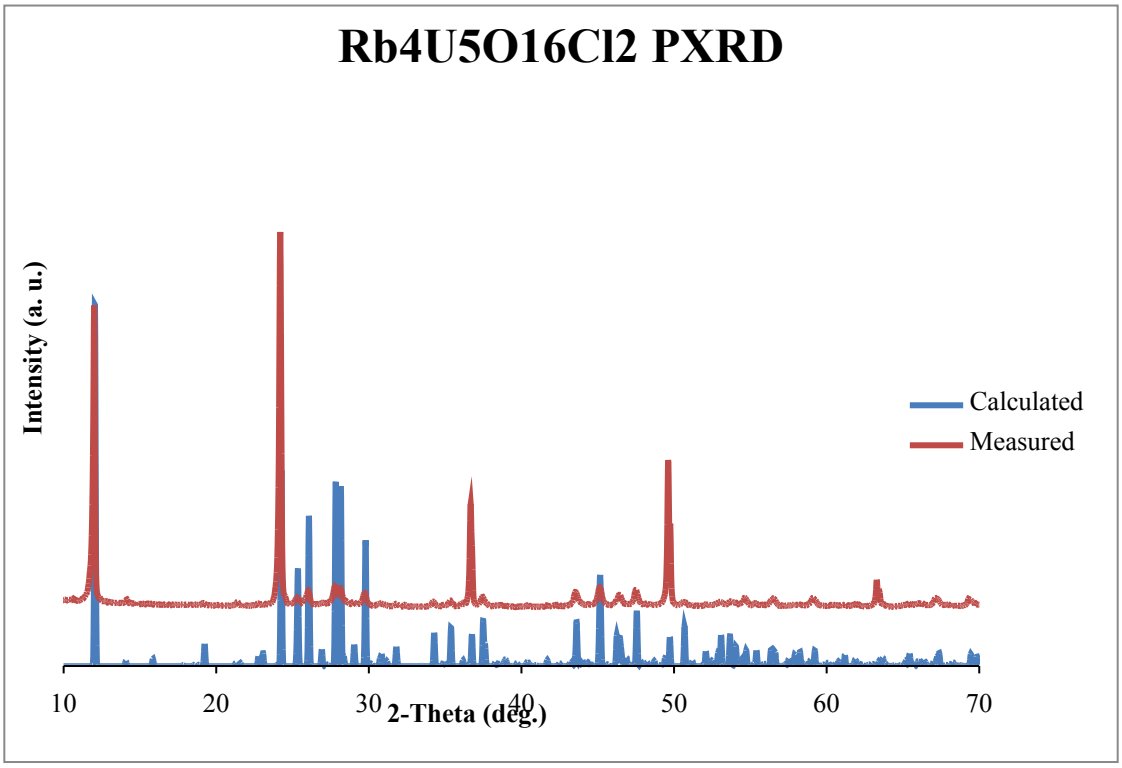

Figure S2: Normalized PXRD patterns for $Rb_4U_5O_{16}Cl_2$, with the measured pattern in red and the calculated pattern in blue.

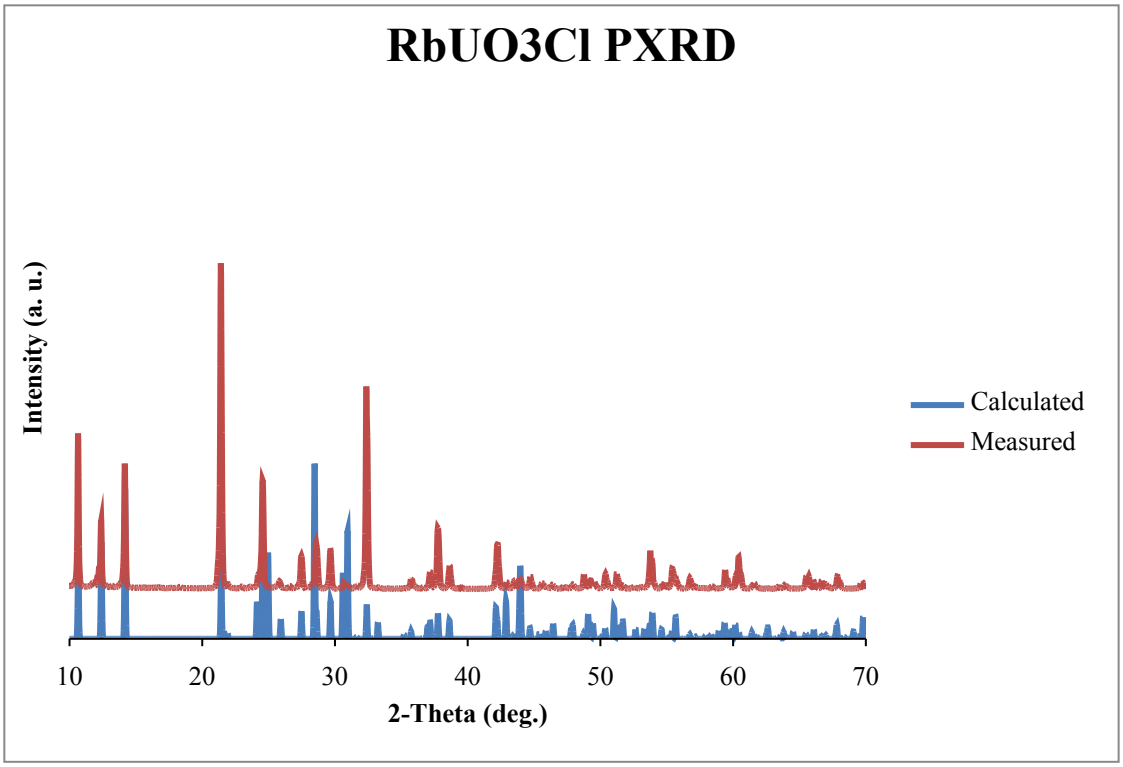

Figure S3: Normalized PXRD patterns for $RbUO_3Cl$, with the measured pattern in red and the calculated pattern in blue.

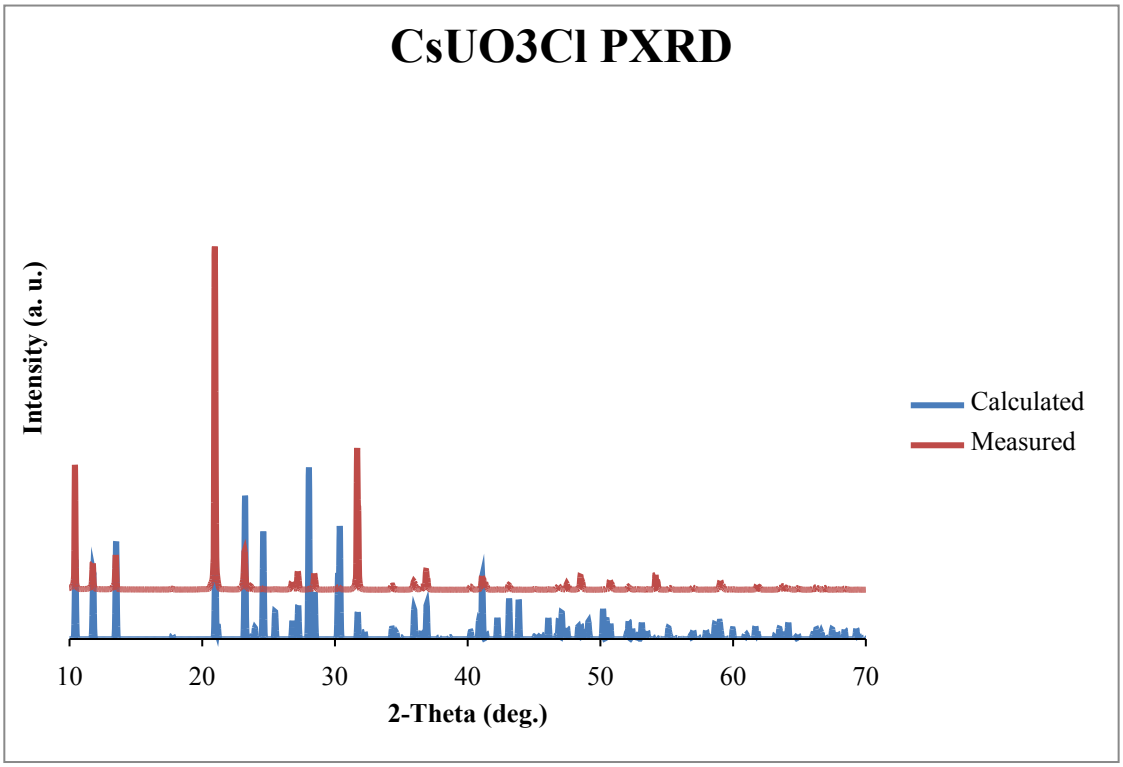

Figure S4: Normalized PXRD patterns for $CsUO_3Cl$, with the measured pattern in red and the calculated pattern in blue.

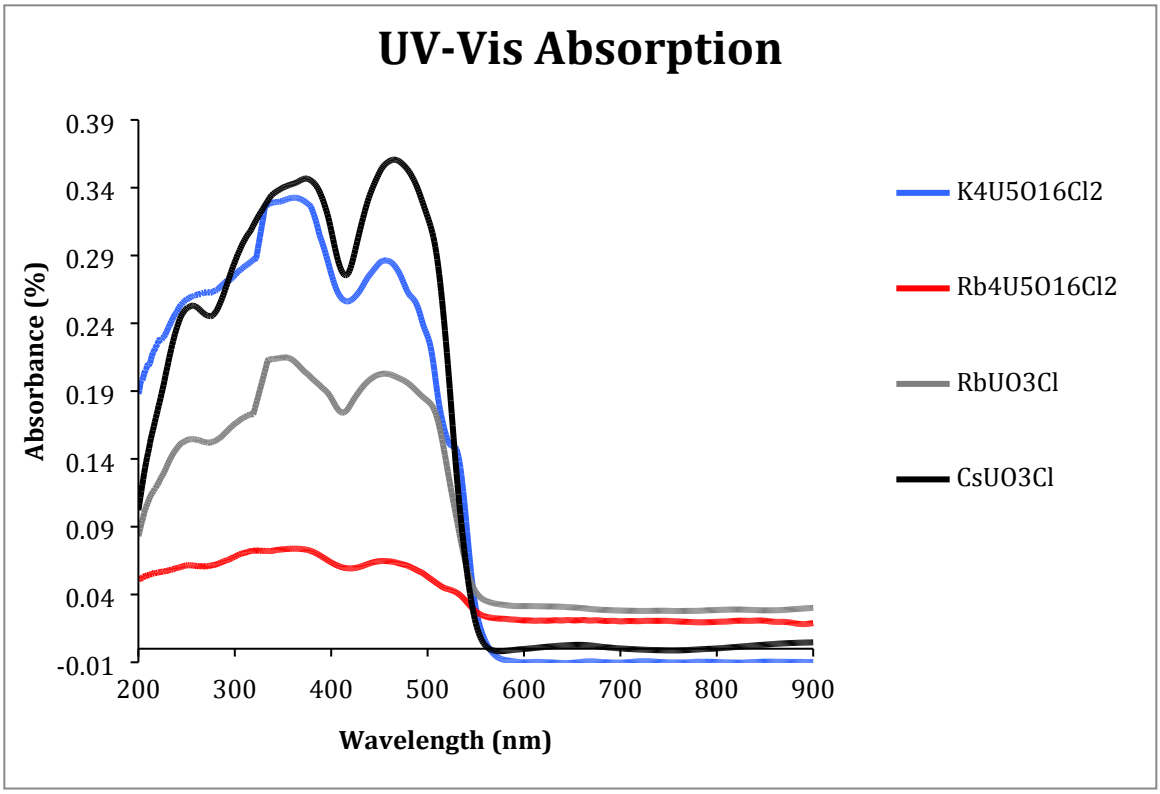

Figure S5: UV-Vis absorbance spectra for $K_4U_5O_{16}Cl_2$ (blue), $Rb_4U_5O_{16}Cl_2$ (red), $RbUO_3Cl$ (grey), and $CsUO_3Cl$ (black).

Table S1. Generalized CCI classification for actinyl ions

CCI Classification Number	Actinyl Donor		Actinyl Acceptor
	a*	***b*****	
0	0	0	0
1	0	1	0
2	1	0	0
3	2	0	0
4	1	1	0
5	0	2	0
6	0	0	1
7	0	1	1
8	1	0	1
9	2	0	1
10	1	1	1
11	0	2	1
12	0	0	2
13	0	1	2
14	1	0	2
15	2	0	2
16	1	1	2
17	0	2	2
18	0	0	3
19	0	1	3
20	1	0	3
21	2	0	3
22	1	1	3
23	0	2	3
24	0	0	4
25	0	1	4
26	1	0	4
27	2	0	4
28	1	1	4
29	0	2	4
30	0	0	5
31	0	1	5
32	1	0	5
33	2	0	5
34	1	1	5
35	0	2	5

*__a__ = two-centered CCI

**__b__ = three-centered CCI

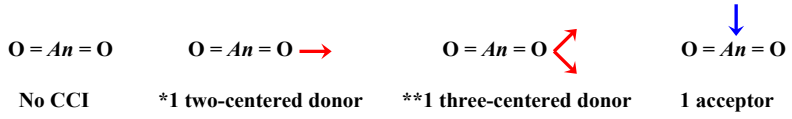

Table S2. Fractional Atomic Coordinates ($\times 10^4$) and Equivalent Isotropic Displacement Parameters ($Å^2 \times 10^3$) for $K_4U_5O_{16}Cl_2$. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	***x***	***y***	***z***	**U(eq)**
U1	7881.61(19)	4847.2(2)	2216.04(13)	11.60(5)
U2	5979.06(19)	4676.8(2)	4040.88(13)	11.80(5)
U3	5000	0	5000	12.26(6)
K1	3096.1(14)	1366(2)	1975.2(10)	26.5(3)
K2	98.2(15)	2648.1(17)	4810.1(10)	27.1(3)
Cl1	2319.7(16)	353(2)	4002.7(12)	29.6(3)
O1	6312(4)	4743(5)	1221(3)	21.1(8)
O2	9479(4)	5010(5)	3200(3)	18.9(8)
O3	4283(4)	4394(6)	3194(3)	20.2(8)
O4	7630(4)	4790(6)	4979(3)	21.5(8)
O5	6707(4)	6733(5)	3140(3)	17.2(7)
O6	6991(4)	2978(5)	3148(3)	18.9(8)
O7	9432(4)	5440(5)	1117(3)	19.8(8)
O8	5075(4)	2979(5)	5196(3)	15.8(7)

Table S3. Fractional Atomic Coordinates ($\times 10^4$) and Equivalent Isotropic Displacement Parameters ($Å^2 \times 10^3$) for $Rb_4U_5O_{16}Cl_2$. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	***x***	***y***	***z***	**U(eq)**
U1	7876.9(2)	4866.5(3)	2227.18(16)	11.90(5)
U2	5988.0(2)	4729.6(3)	4044.70(15)	12.28(5)
U3	5000	0	5000	12.69(7)
Rb1	3092.8(7)	1320.5(10)	2005.5(5)	26.13(14)
Rb2	40.1(7)	2627.4(8)	4837.2(5)	25.68(14)
Cl1	2382.8(18)	319(3)	4070.8(14)	29.0(4)
O1	6360(5)	4732(6)	1289(4)	21.3(9)
O2	9419(5)	5041(6)	3173(4)	22(1)
O3	4388(5)	4445(6)	3204(3)	21.6(9)
O4	7554(5)	4848(6)	4968(4)	21.0(9)
O5	6737(4)	6749(6)	3139(3)	16.7(8)
O6	7008(4)	2978(6)	3171(3)	18.5(9)
O7	9447(5)	5363(6)	1128(3)	20.8(9)
O8	5069(4)	2979(6)	5172(3)	16.2(9)

Table S4. Fractional Atomic Coordinates ($\times 10^4$) and Equivalent Isotropic Displacement Parameters ($Å^2 \times 10^3$) for $Cs_5U_7O_{22}Cl_3$. U_{eq} is defined as 1/3 of of the trace of the orthogonalised U_{IJ} tensor.

Atom	*x*	*y*	*z*	U(eq)
U1	1091.9(3)	2192.04(18)	5105.5(3)	15.87(8)
U2	1133.8(3)	4238.31(18)	5112.6(3)	15.66(8)
U3	3345.8(3)	642.83(19)	5017.7(3)	15.68(8)
U4	4292.5(3)	3001.52(19)	5103.8(3)	15.45(8)
U5	7023.0(3)	1421.91(18)	5018.1(3)	16.32(8)
U6	7923.7(3)	3447.50(19)	5060.9(3)	15.81(8)
U7	0	0	5000	16.77(11)
U8	5000	5000	5000	16.51(11)
Cs1	2368.6(7)	1959.0(4)	2761.3(5)	28.83(16)
Cs2	5202.1(7)	3553.4(5)	2544.4(6)	36.78(19)
Cs3	4333.9(7)	244.5(4)	2341.6(5)	30.07(16)
Cs4	9114.5(7)	3234.0(4)	2506.9(5)	29.39(16)
Cs5	8804.8(8)	1049.5(4)	2534.1(6)	37.18(19)
Cl1	6302(3)	1630.8(17)	3312(2)	37.0(7)
Cl2	7059(3)	1361.3(16)	6702(2)	30.8(6)
Cl3	4020(3)	5084.3(16)	3301(2)	33.3(7)
O1	604(7)	2236(4)	3949(5)	25.2(17)

O2	1515(7)	2086(4)	6256(5)	25.9(17)
O3	1246(6)	959(3)	4957(6)	23.6(18)
O4	3310(7)	1868(3)	5048(5)	21.8(17)
O5	2353(6)	3190(3)	5243(5)	21.8(17)
O6	-66(6)	3204(3)	5184(5)	20.8(16)
O7	-1381(7)	1927(4)	5117(5)	24.3(17)
O8	784(8)	4117(4)	3974(6)	28.1(18)
O9	1513(8)	4375(4)	6262(6)	28.1(18)
O10	3553(7)	4445(4)	5139(5)	22.7(16)
O11	1043(7)	5470(4)	4914(6)	24.8(19)
O12	3006(7)	603(4)	3873(5)	23.8(17)
O13	3774(8)	584(4)	6177(6)	27.8(18)
O14	2055(7)	-465(4)	5051(6)	26.6(19)
O15	4436(7)	-743(4)	5062(5)	20.0(15)
O16	3857(7)	3046(4)	3963(6)	26.4(18)
O17	4834(7)	2990(4)	6257(5)	24.2(17)
O18	6256(6)	2431(3)	5037(5)	17.3(15)
O19	7609(7)	3409(4)	3907(6)	27.5(18)
O20	8141(7)	3445(4)	6208(5)	25.8(17)
O21	343(8)	93(4)	6150(5)	27.6(18)
O22	4152(6)	6014(4)	5058(5)	20.5(16)

Table S5. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for $RbUO_3Cl$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
U(1)	3798(1)	2500	3247(1)	12(1)
Rb(1)	1458(2)	2500	7496(2)	25(1)
Cl(1)	7825(4)	2500	9221(4)	22(1)
O(1)	5511(12)	2500	5847(11)	22(2)
O(2)	1592(12)	2500	3816(11)	21(2)
O(3)	5904(12)	2500	2490(11)	23(2)

Table S6. Atomic coordinates (x 10^4) and equivalent isotropic displacement parameters ($Å^2$x 10^3) for $CsUO_3Cl$. U(eq) is defined as one third of the trace of the orthogonalized U^{ij} tensor.

	x	y	z	U(eq)
U(1)	3802(1)	2500	3295(1)	18(1)
Cs(1)	1498(1)	2500	7524(1)	29(1)
Cl(1)	7827(3)	2500	9113(2)	28(1)
O(1)	5535(8)	2500	5825(7)	27(1)
O(2)	1740(8)	2500	3883(8)	29(1)
O(3)	5708(8)	2500	2493(8)	32(1)