

Table S-A. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0$

	T = 90 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
Formula	$CuH_{12}O_{14}Rb_2Se_2$	$CuH_{12}O_{14}Rb_2Se_2$	$CuH_{12}O_{14}Rb_2Se_2$	$CuH_{12}O_{14}Rb_2Se_2$	$CuH_{12}O_{14}Rb_2Se_2$
fw ^a	628.50	628.50	628.50	628.50	628.50
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.3811(2)	9.3851(2)	9.3926(2)	9.3974(2)	9.3982(2)
b (Å)	12.4639(4)	12.4788(4)	12.4986(4)	12.5143(4)	12.5275(4)
c (Å)	6.3211(2)	6.3235(2)	6.3290(2)	6.3347(2)	6.3404(2)
β° (deg)	104.566(2)	104.573(2)	104.598(2)	104.625(2)	104.657(2)
V (Å ³)	715.34(3)	716.75(3)	719.00(3)	720.83(3)	722.20(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.918	2.912	2.903	2.895	2.890
occupancy x	0	0	0	0	0
F(000) ^a	590	590	590	590	590
abs. coeff. (mm ⁻¹)	13.43	13.40	13.36	13.32	13.30
2 θ range (deg)	3.2-81.7	3.2-80.0	3.2-74.0	3.2-78.7	3.2-80.4
index range h, k, l	-17 17, -22 22, -11 11	-16 16, -22 22, -11 11	-15 15, -21 20, -10 10	-16 16, -22 21, -11 11	-16 16, -22 22, -11 11
no. refl. Collected	9001	8712	6886	8167	8718
no. unique refl.	4594	4391	3614	4195	4442
R equivalent refl.	0.028	0.025	0.029	0.028	0.036
data completeness	0.984	0.990	0.991	0.979	0.982
GOF ^b on F	1.28	1.42	1.29	1.30	1.26
no. refl. > 5 σ (I)	3004	2941	2183	2451	2188
R_1 [$I > 5\sigma(I)$] ^c	0.022	0.022	0.021	0.020	0.021
w R_2 [$I > 5\sigma(I)$] ^d	0.023	0.022	0.020	0.021	0.021
extinct. coeff.	1.171×10^{-6}	1.244×10^{-6}	1.005×10^{-6}	1.161×10^{-6}	1.202×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.66/-1.24	0.66/-1.15	0.62/-0.76	0.48/-0.51	0.47/-0.52

Table S-A. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0$

	T = 270 K	T = 298 K
formula	$CuH_{12}O_{14}Rb_2Se_2$	$CuH_{12}O_{14}Rb_2Se_2$
fw ^a	628.50	628.50
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$
a (Å)	9.3991(2)	9.3985(2)
b (Å)	12.5403(4)	12.5560(3)
c (Å)	6.3465(2)	6.3540(2)
β° (deg)	104.683(1)	104.716(1)
V (Å ³)	723.62(3)	725.22(3)
Z	2	2
d _{calc} (g cm ⁻³) ^a	2.884	2.878
occupancy x	0	0
F(000) ^a	590	590
abs. coeff. (mm ⁻¹)	13.27	13.24
2 θ range (deg)	3.2-80.5	3.2-72.6
index range <i>h, k, l</i>	-16 16, -22 21, -11 11	-15 15, -20 20, -10 10
no. refl. collected	8677	6741
no. unique refl.	4457	3436
R equivalent refl.	0.030	0.022
data completeness	0.983	0.982
GOF ^b on F	1.37	1.54
no. refl. > 5 σ (I)	2245	2150
R ₁ [I > 5 σ (I)] ^c	0.020	0.020
wR ₂ [I > 5 σ (I)] ^d	0.021	0.022
extinct. coeff.	1.271×10^{-6}	1.597×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.59/-0.45	0.60/-0.58

Table S-B. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.127$

	T = 90 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
formula	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$
fw ^a	616.71	616.71	616.71	616.71	616.71
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	Monoclinic	monoclinic
space group	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a
a (Å)	9.3532(3)	9.3578(3)	9.3636(2)	9.3683(3)	9.3688(3)
b (Å)	12.4355(3)	12.4490(3)	12.4659(2)	12.4828(4)	12.4969(4)
c (Å)	6.3154(2)	6.3189(2)	6.3238(1)	6.3321(2)	6.3379(2)
β° (deg)	104.496(1)	104.518(1)	104.549(1)	104.586(2)	104.615(2)
V (Å ³)	711.17(3)	712.62(3)	714.48(2)	716.63(4)	718.04(4)
Z	2	2	2	2	2
d _{calc} (g cm ⁻³) ^a	2.880	2.874	2.866	2.858	2.852
occupancy x	0.124(1)	0.134(1)	0.130(1)	0.125(1)	0.126(1)
F(000) ^a	580.9	580.9	580.9	580.9	580.9
abs. coeff. (mm ⁻¹)	12.74	12.65	12.64	12.63	12.60
2θ range (deg)	3.2-69.9	3.2-67.4	3.2-75.6	3.2-67.4	3.2-68.9
index range <i>h, k, l</i>	-15 15, -19 19, -10 10	-14 14, -19 14, -9 9	-16 16, -19 21, -10 10	-14 14, -19 17, -9 9	-14 14, -19 17, -10 10
no. refl. collected	5972	5193	7482	5565	5750
no. unique refl.	3106	2835	3804	2852	2984
R equivalent refl.	0.024	0.021	0.024	0.035	0.033
data completeness	0.997	0.998	0.995	0.998	0.999
GOF ^b on F	1.58	1.69	1.53	1.52	1.59
no. refl. > 5σ(I)	2349	2205	2594	1810	1820
R ₁ [I > 5σ(I)] ^c	0.024	0.025	0.022	0.028	0.030
wR ₂ [I > 5σ(I)] ^d	0.026	0.027	0.021	0.028	0.030
extinct. coeff.	7.091×10 ⁻⁷	7.370×10 ⁻⁷	8.300×10 ⁻⁷	7.412×10 ⁻⁷	8.487×10 ⁻⁷
largest diff. peak and hole (e Å ⁻³)	1.22/-1.72	1.09/-1.73	0.67/-1.02	1.05/-1.12	1.05/-1.08

Table S-B. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.127$

	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$	$CuH_{12}K_{0.254}O_{14}Rb_{1.746}Se_2$
fw ^a	616.71	616.71
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	Monoclinic
space group	$P2_1/a$	$P2_1/a$
a (Å)	9.3700(2)	9.3660(4)
b (Å)	12.5081(2)	12.5179(5)
c (Å)	6.3463(1)	6.3535(3)
β° (deg)	104.622(1)	104.641(2)
V (Å ³)	719.70(2)	720.71(5)
Z	2	2
d_{calc} (g cm ⁻³) ^a	2.846	2.842
occupancy x	0.1289(8)	0.123(2)
F(000) ^a	580.9	580.9
abs. coeff. (mm ⁻¹)	12.55	12.57
2 θ range (deg)	3.2-75.6	3.2-67.4
index range h, k, l	-16 16, -21 20, -10 10	-14 14, -19 16, -9 9
no. refl. collected	7455	4756
no. unique refl.	3832	2866
R equivalent refl.	0.024	0.031
data completeness	0.996	0.998
GOF ^b on F	1.62	1.47
No. refl. > 5 σ (I)	2329	1577
R_1 [$I > 5\sigma(I)$] ^c	0.023	0.028
wR_2 [$I > 5\sigma(I)$] ^d	0.023	0.029
extinct. coeff.	9.725×10^{-7}	7.300×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.69/-0.86	0.91/-0.82

Table S-C. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.262$

	T = 85 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
formula	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$
fw ^a	604.19	604.19	604.19	604.19	604.19
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	Monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.3342(3)	9.3295(3)	9.3381(3)	9.3399(3)	9.3391(3)
b (Å)	12.4193(3)	12.4126(3)	12.4331(3)	12.4495(3)	12.4605(3)
c (Å)	6.3112(2)	6.3095(2)	6.3163(2)	6.3246(2)	6.3328(2)
β° (deg)	104.419(1)	104.419(1)	104.445(1)	104.466(1)	104.485(2)
V (Å ³)	708.58(3)	707.65(3)	710.15(3)	712.09(3)	713.52(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.832	2.835	2.825	2.818	2.812
occupancy x	0.2594(9)	0.267(2)	0.263(1)	0.2598(8)	0.2617(9)
F(000) ^a	571.1	571.1	571.1	571.1	571.1
abs. coeff. (mm ⁻¹)	11.94	11.91	11.89	11.88	11.84
2 θ range (deg)	3.2-75.5	3.2-84.3	3.2-78.8	3.2-80.6	3.2-74.1
index range h, k, l	-16 16, -21 21, -10 10	-17 17, -22 23, -11 11	-16 16, -21 16, -11 11	-16 16, -22 22, -11 11	-15 15, -20 20, -10 10
no. refl. collected	7513	9037	7353	8645	7115
no. unique refl.	3766	4891	4153	4414	3612
R equivalent refl.	0.026	0.026	0.029	0.028	0.026
data completeness	0.998	0.979	0.978	0.985	0.992
GOF ^b on F	1.34	1.20	1.24	1.24	1.24
no. refl. > 5 σ (I)	2450	2929	2381	2468	2118
R ₁ [$I > 5\sigma(I)$] ^c	0.020	0.021	0.021	0.019	0.018
wR ₂ [$I > 5\sigma(I)$] ^d	0.021	0.021	0.022	0.019	0.019
extinct. coeff.	4.400×10^{-7}	4.985×10^{-7}	3.619×10^{-7}	4.752×10^{-7}	5.644×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.51/-0.71	0.48/-0.56	0.54/-0.47	0.41/-0.58	0.40/-0.51

Table S-C. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.262$

	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$	$CuH_{12}K_{0.524}O_{14}Rb_{1.476}Se_2$
fw ^a	604.19	604.19
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$
a (Å)	9.3369(3)	9.3336(3)
b (Å)	12.4707(3)	12.4775(4)
c (Å)	6.3437(2)	6.3543(2)
β° (deg)	104.495(1)	104.490(2)
V (Å ³)	715.13(3)	716.48(4)
Z	2	2
d_{calc} (g cm ⁻³) ^a	2.806	2.800
occupancy x	0.260(1)	0.2635(9)
F(000) ^a	571.1	571.1
abs. coeff. (mm ⁻¹)	11.83	11.78
2 θ range (deg)	3.2-74.2	3.2-74.0
index range <i>h, k, l</i>	-15 15, -20 21, -10 10	-15 15, -15 21, -10 10
no. refl. collected	7225	5984
no. unique refl.	3625	3594
R equivalent refl.	0.025	0.024
data completeness	0.994	0.987
GOF ^b on F	1.33	1.54
no. refl. > 5 σ (I)	2069	2036
R_1 [$I > 5\sigma(I)$] ^c	0.018	0.021
wR_2 [$I > 5\sigma(I)$] ^d	0.019	0.022
extinct. coeff.	5.939×10^{-7}	8.373×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.40/-0.48	0.43/-0.46

Table S-D. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.318$

	T = 90 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
formula	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$
fw ^a	599.00	599.00	599.00	599.00	599.00
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	Monoclinic	monoclinic
space group	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a
a (Å)	9.3184(2)	9.3230(2)	9.3262(2)	9.3262(2)	9.3221(2)
b (Å)	12.3936(2)	12.4093(4)	12.4237(4)	12.4351(4)	12.4440(3)
c (Å)	6.3027(1)	6.3082(2)	6.3148(2)	6.3239(2)	6.3348(1)
β° (deg)	104.3747(9)	104.402(2)	104.426(2)	104.441(2)	104.429(1)
V (Å ³)	705.10(2)	706.87(3)	708.60(3)	710.23(3)	711.68(2)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.821	2.814	2.807	2.801	2.795
occupancy x	0.3180(7)	0.314(1)	0.318(1)	0.3149(9)	0.3214(8)
F(000) ^a	567.1	567.1	567.1	567.1	567.1
abs. coeff. (mm ⁻¹)	11.63	11.63	11.57	11.57	11.50
2 θ range (deg)	3.2-72.6	3.2-67.4	3.2-67.4	3.2-68.7	3.2-71.2
index range h, k, l	-15 15, -20 20, -10 10	-14 14, -19 19, -9 9	-14 14, -19 19, -9 9	-14 14, -19 19, -10 10	-15 15, -20 20, -10 10
no. refl. collected	6600	5530	5650	5899	6434
no. unique refl.	3336	2805	2824	2948	3232
R equivalent refl.	0.019	0.020	0.019	0.019	0.018
data completeness	0.970	0.997	0.999	0.998	0.986
GOF ^b on F	1.90	1.63	1.69	1.68	1.98
no. refl. > 5 σ (I)	2713	2191	2170	2171	2280
R ₁ [$I > 5\sigma(I)$] ^c	0.020	0.021	0.019	0.020	0.021
wR ₂ [$I > 5\sigma(I)$] ^d	0.022	0.024	0.023	0.023	0.023
extinct. coeff.	1.281×10^{-6}	1.208×10^{-6}	1.231×10^{-6}	1.315×10^{-6}	1.205×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.62/-0.84	0.71/-0.60	0.60/-0.52	0.57/-0.46	0.54/-0.58

Table S-D. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.318$

	T = 270 K	T = 298 K	T = 310 K	T = 315 K
formula	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$	$CuH_{12}K_{0.636}O_{14}Rb_{1.364}Se_2$
fw ^a	599.00	599.00	599.00	599.00
wavelength (Å)	0.71069	0.71069	0.71069	0.71069
crystal system	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.3171(2)	9.3153(2)	9.3154(2)	9.3158(3)
b (Å)	12.4522(4)	12.4599(3)	12.4668(5)	12.4672(6)
c (Å)	6.3472(2)	6.3572(2)	6.3599(3)	6.3623(3)
β° (deg)	104.436(2)	104.431(1)	104.445(2)	104.443(3)
V (Å ³)	713.14(3)	714.58(3)	715.25(4)	715.58(5)
Z	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.789	2.784	2.781	2.780
occupancy x	0.321(1)	0.3188(7)	0.316(1)	0.318(1)
F(000) ^a	567.1	567.1	567.1	567.1
abs. coeff. (mm ⁻¹)	11.48	11.47	11.48	11.46
2 θ range (deg)	3.2-68.7	3.2-75.6	3.2-67.4	3.2-67.6
index range h, k, l	-14 14, -19 19, -10 10	-15 15, -21 20, -10 10	-14 14, -17 19, -9 9	-14 14, -19 16, -9 9
no. refl. collected	5927	7474	5284	5222
no. unique refl.	2967	3801	2856	2861
R equivalent refl.	0.020	0.025	0.028	0.033
data completeness	0.998	0.998	0.998	0.996
GOF ^b on F	1.77	1.70	1.47	1.50
no. refl. > 5 σ (I)	2009	2245	1765	1653
R_1 [$I > 5\sigma(I)$] ^c	0.022	0.022	0.023	0.026
wR_2 [$I > 5\sigma(I)$] ^d	0.024	0.023	0.025	0.027
extinct. coeff.	1.342×10^{-6}	1.379×10^{-6}	1.141×10^{-6}	1.221×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.52/-0.49	0.64/-0.74	0.44/-0.57	0.65/-0.75

Table S-E. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.359$

	T = 90 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
formula	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$
fw ^a	595.20	595.20	595.20	595.20	595.20
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	Monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.3092(2)	9.3121(2)	9.3147(2)	9.3140(2)	9.3094(2)
b (Å)	12.3748(3)	12.3865(5)	12.4000(4)	12.4145(4)	12.4242(3)
c (Å)	6.2996(2)	6.3033(2)	6.3096(2)	6.3209(2)	6.3357(1)
β° (deg)	104.334(1)	104.360(2)	104.381(2)	104.396(2)	104.375(1)
V (Å ³)	703.12(3)	704.33(4)	705.94(3)	707.93(3)	709.86(2)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.811	2.806	2.800	2.792	2.784
occupancy x	0.3556(8)	0.3599(9)	0.357(1)	0.357(1)	0.362(2)
F(000) ^a	564.2	564.2	564.2	564.2	564.2
abs. coeff. (mm ⁻¹)	11.43	11.38	11.37	11.34	11.28
2 θ range (deg)	3.2-69.9	3.2-68.7	3.2-67.4	3.2-67.4	3.2-75.6
index range h, k, l	-14 14, -17 19, -10 10	-14 14, -13 19, -9 10	-14 14, -17 19, -9 9	-14 14, -17 19, -9 9	-15 15, -21 21, -10 10
no. refl. collected	5677	4575	5236	5198	7140
no. unique refl.	3080	2935	2807	2813	3664
R equivalent refl.	0.020	0.021	0.021	0.024	0.024
data completeness	0.998	0.999	0.999	0.998	0.967
GOF ^b on F	1.33	1.37	1.33	1.51	1.71
no. refl. > 5 σ (I)	2421	2210	2094	1967	2278
R_1 [$I > 5\sigma(I)$] ^c	0.018	0.020	0.019	0.022	0.021
wR_2 [$I > 5\sigma(I)$] ^d	0.020	0.024	0.023	0.026	0.024
extinct. coeff.	1.226×10^{-6}	1.072×10^{-6}	1.350×10^{-6}	1.298×10^{-6}	1.310×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.64/-0.52	0.48/-0.61	0.53/-0.54	0.55/-0.61	0.72/-0.62

Table S-E. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.359$

	T = 255 K	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$	$CuH_{12}K_{0.718}O_{14}Rb_{1.282}Se_2$
fw ^a	595.20	595.20	595.20
wavelength (Å)	0.71069	0.71069	0.71069
crystal system	monoclinic	monoclinic	monoclinic
space group	P2 ₁ /a	P2 ₁ /a	P2 ₁ /a
a (Å)	9.3017(2)	9.3002(1)	9.3021(2)
b (Å)	12.4232(4)	12.4295(2)	12.4428(4)
c (Å)	6.3417(2)	6.3517(1)	6.3617(2)
β° (deg)	104.379(2)	104.3582(9)	104.361(2)
V (Å ³)	709.87(3)	711.30(2)	713.32(3)
Z	2	2	2
d_{calc} (g cm ⁻³) ^a	2.784	2.779	2.771
occupancy x	0.358(1)	0.3672(7)	0.359(1)
F(000) ^a	564.2	564.2	564.2
abs. coeff. (mm ⁻¹)	11.31	11.23	11.24
2 θ range (deg)	3.2-68.7	3.2-75.6	3.2-68.6
index range h, k, l	-14 14, -19 19, -10 10	-15 15, -21 21, -10 10	-14 14, -16 19, -10 10
no. refl. collected	5478	7209	5312
no. unique refl.	2951	3704	2977
R equivalent refl.	0.024	0.020	0.023
data completeness	0.998	0.977	0.999
GOF ^b on F	1.53	1.61	1.33
no. refl. > 5 σ (I)	1925	2155	1850
R ₁ [I > 5 σ (I)] ^c	0.023	0.019	0.021
wR ₂ [I > 5 σ (I)] ^d	0.026	0.019	0.023
extinct. coeff.	1.103×10^{-6}	1.941×10^{-6}	1.346×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.59/-0.40	0.53/-0.47	0.43/-0.43

Table S-F. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.390$

	T = 90 K	T = 130 K	T = 170 K	T = 190 K	T = 210 K
formula	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$
fw ^a	592.32	592.32	592.32	592.32	592.32
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.3054(4)	9.3083(3)	9.3089(3)	9.3084(3)	9.3032(3)
b (Å)	12.3738(5)	12.3865(4)	12.3982(4)	12.4065(5)	12.4100(5)
c (Å)	6.2993(2)	6.3022(2)	6.3087(1)	6.3159(1)	6.3230(2)
β° (deg)	104.337(2)	104.344(2)	104.356(2)	104.357(2)	104.351(2)
V (Å ³)	702.73(5)	703.97(4)	705.37(3)	706.61(3)	707.23(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.799	2.794	2.789	2.784	2.781
occupancy x	0.3973(8)	0.390(1)	0.3902(7)	0.3859(9)	0.3937(8)
F(000) ^a	561.9	561.9	561.9	561.9	561.9
abs. coeff. (mm ⁻¹)	11.17	11.20	11.18	11.18	11.12
2 θ range (deg)	3.2-86.2	3.2-84.2	3.2-82.3	3.2-75.5	3.2-74.0
index range h, k, l	-17 17, -23 23, -12 12	-17 17, -23 23, -11 11	-17 17, -22 22, -11 11	-16 16, -21 21, -10 10	-15 15, -20 21, -10 10
no. refl. collected	10002	9528	9017	7209	6875
no. unique refl.	5108	4833	4612	3739	3553
R equivalent refl.	0.030	0.031	0.027	0.026	0.026
data completeness	0.981	0.975	0.985	0.996	0.992
GOF ^b on F	1.27	1.26	1.36	1.38	1.65
no. refl. > 5 σ (I)	3294	2912	2749	2288	2163
R_1 [$I > 5\sigma(I)$] ^c	0.022	0.021	0.021	0.022	0.024
wR_2 [$I > 5\sigma(I)$] ^d	0.023	0.021	0.021	0.022	0.026
extinct. coeff.	1.447×10^{-6}	1.081×10^{-6}	1.086×10^{-6}	1.051×10^{-6}	1.012×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.67/-0.69	0.43/-0.58	0.45/-0.85	0.58/-0.64	0.64/-1.20

Table S-F. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.390$

	T = 225 K	T = 230 K	T = 235 K	T = 240 K	T = 255 K
formula	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$
fw ^a	592.32	592.32	592.32	592.32	592.32
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	Monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.2933(3)	9.2900(4)	9.2822(3)	9.2810(3)	9.2809(3)
b (Å)	12.4104(5)	12.4107(5)	12.4069(5)	12.4109(5)	12.4147(4)
c (Å)	6.3347(2)	6.3395(2)	6.3455(2)	6.3544(1)	6.3593(1)
β° (deg)	104.314(2)	104.306(2)	104.282(2)	104.261(2)	104.258(2)
V (Å ³)	707.92(4)	708.25(4)	708.18(4)	709.38(3)	710.14(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.779	2.777	2.778	2.773	2.770
occupancy x	0.391(1)	0.385(1)	0.393(1)	0.3858(8)	0.391(1)
F(000) ^a	561.9	561.9	561.9	561.9	561.9
abs. coeff. (mm ⁻¹)	11.13	11.16	11.11	11.14	11.10
2 θ range (deg)	3.2-75.5	3.2-75.6	3.2-75.6	3.2-74.1	3.2-74.1
index range h, k, l	-16 15, -21 21, -10 10	-16 15, -21 21, -10 10	-16 15, -21 21, -10 10	-15 15, -21 21, -10 10	-15 15, -20 21, -10 10
no. refl. collected	7332	7215	7260	6884	7102
no. unique refl.	3750	3749	3748	3572	3574
R equivalent refl.	0.026	0.029	0.029	0.023	0.024
data completeness	0.995	0.992	0.993	0.990	0.990
GOF ^b on F	1.81	1.69	1.68	1.65	1.43
no. refl. > 5 σ (I)	2247	2095	2136	2186	2142
R_1 [$I > 5\sigma(I)$] ^c	0.028	0.026	0.026	0.024	0.021
wR_2 [$I > 5\sigma(I)$] ^d	0.028	0.027	0.027	0.025	0.021
extinct. coeff.	1.026×10^{-6}	9.871×10^{-7}	8.850×10^{-7}	1.088×10^{-6}	1.043×10^{-6}
largest diff. peak and hole (e Å ⁻³)	1.21/-1.12	0.64/-0.67	0.78/-1.07	0.55/-0.69	0.42/-0.57

Table S-F. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.390$

	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$	$CuH_{12}K_{0.780}O_{14}Rb_{1.220}Se_2$
fw ^a	592.32	592.32
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$
a (Å)	9.2844(3)	9.2922(2)
b (Å)	12.4217(5)	12.4333(3)
c (Å)	6.3620(2)	6.3652(7)
β° (deg)	104.265(2)	104.295(1)
V (Å ³)	711.09(4)	712.62(3)
Z	2	2
d_{calc} (g cm ⁻³) ^a	2.766	2.760
occupancy x	0.3852(8)	0.3918(7)
F(000) ^a	561.9	561.9
abs. coeff. (mm ⁻¹)	11.12	11.05
2 θ range (deg)	3.2-78.8	3.2-75.6
index range h, k, l	-16 16, -22 22, -11 11	-16 15, -21 21, -10 10
no. refl. collected	8146	7406
no. unique refl.	4135	3770
R equivalent refl.	0.034	0.025
data completeness	0.975	0.990
GOF ^b on F	1.34	1.41
no. refl. > 5 σ (I)	1979	2122
R_1 [$I > 5\sigma(I)$] ^c	0.020	0.019
wR_2 [$I > 5\sigma(I)$] ^d	0.020	0.020
extinct. coeff.	1.031×10^{-6}	1.170×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.37/-0.46	0.36/-0.41

Table S-G. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.411$

	T = 90 K	T = 130 K	T = 170 K	T = 190 K	T = 200 K
formula	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$
fw ^a	590.38	590.38	590.38	590.38	590.38
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.2979(2)	9.3035(2)	9.3070(2)	9.3055(2)	9.2928(3)
b (Å)	12.3626(3)	12.3798(3)	12.3932(3)	12.4003(3)	12.3922(4)
c (Å)	6.2966(2)	6.3023(2)	6.3092(2)	6.3154(2)	6.3269(2)
β° (deg)	104.310(1)	104.329(1)	104.346(1)	104.339(1)	104.297(2)
V (Å ³)	701.31(3)	703.29(3)	705.03(3)	706.04(3)	706.03(4)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.796	2.788	2.781	2.777	2.777
occupancy x	0.4155(8)	0.410(1)	0.4127(7)	0.4106(8)	0.410(2)
F(000) ^a	560.4	560.4	560.4	560.4	560.4
abs. coeff. (mm ⁻¹)	11.08	11.08	11.04	11.04	11.04
2 θ range (deg)	3.2-80.5	3.2-72.6	3.2-74.2	3.2-74.1	3.2-72.8
index range h, k, l	-16 16, -22 21, -11 11	-15 15, -20 20, -10 10	-15 15, -16 20, -10 10	-15 15, -20 20, -10 10	-15 15, -20 19, -10 10
no. refl. collected	8509	6705	6714	7072	6665
no. unique refl.	4352	3382	3580	3573	3375
R equivalent refl.	0.025	0.020	0.021	0.022	0.029
data completeness	0.988	0.990	0.993	0.996	0.981
GOF ^b on F	1.28	1.36	1.39	1.61	2.45
no. refl. > 5 σ (I)	2945	2521	2530	2462	2121
R ₁ [$I > 5\sigma(I)$] ^c	0.020	0.019	0.020	0.021	0.038
wR ₂ [$I > 5\sigma(I)$] ^d	0.021	0.021	0.021	0.024	0.039
extinct. coeff.	1.315×10^{-6}	1.539×10^{-6}	1.448×10^{-6}	1.425×10^{-6}	9.006×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.73/-0.67	0.46/-0.75	0.44/-0.85	0.47/-0.78	1.17/-1.19

Table S-G. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.411$

	T = 210 K	T = 240 K	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$	$CuH_{12}K_{0.822}O_{14}Rb_{1.178}Se_2$
fw ^a	590.38	590.38	590.38	590.38
wavelength (Å)	0.71069	0.71069	0.71069	0.71069
crystal system	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.2511(3)	9.2644(2)	9.2763(2)	9.2833(2)
b (Å)	12.3828(3)	12.3968(3)	12.4118(3)	12.4218(3)
c (Å)	6.3675(2)	6.3637(2)	6.3643(2)	6.3657(2)
β° (deg)	104.071(1)	104.160(1)	104.220(1)	104.252(1)
V (Å ³)	707.54(3)	708.66(3)	710.30(3)	711.47(3)
Z	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.771	2.767	2.760	2.756
occupancy x	0.412(1)	0.4079(8)	0.414(1)	0.405(2)
F(000) ^a	560.4	560.4	560.4	560.4
abs. coeff. (mm ⁻¹)	11.01	11.01	10.95	10.99
2 θ range (deg)	3.2-75.5	3.2-72.6	3.2-72.6	3.2-75.6
index range h, k, l	-15 15, -20 21, -10 10	-15 15, -20 19, -10 10	-15 15, -20 19, -10 10	-15 15, -21 20, -10 10
no. refl. collected	7518	6732	6751	7269
no. unique refl.	3759	3421	3430	3760
R equivalent refl.	0024	0.022	0.020	0.023
data completeness	0.999	0.992	0.992	0.991
GOF ^b on F	1.43	1.44	1.39	1.40
no. refl. > 5 σ (I)	2346	2247	2273	2203
R_1 [$I > 5\sigma(I)$] ^c	0.020	0.019	0.019	0.018
wR_2 [$I > 5\sigma(I)$] ^d	0.022	0.021	0.020	0.020
extinct. coeff.	1.202×10^{-6}	1.383×10^{-6}	1.414×10^{-6}	1.556×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.46/-0.57	0.43/-0.49	0.35/-0.48	0.33/-0.36

Table S-H. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.498$

	T = 90 K	T = 130 K	T = 170 K	T = 210 K	T = 240 K
formula	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$
fw ^a	582.31	582.31	582.31	582.31	582.31
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.2009(2)	9.2132(2)	9.2205(2)	9.2356(4)	9.2450(2)
b (Å)	12.3225(3)	12.3355(3)	12.3439(3)	12.3614(4)	12.3730(3)
c (Å)	6.3582(2)	6.3628(2)	6.3635(2)	6.3658(3)	6.3648(2)
β° (deg)	103.729(1)	103.783(1)	103.853(1)	103.935(3)	104.009(1)
V (Å ³)	700.28(3)	702.30(3)	703.21(3)	705.36(5)	706.41(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.761	2.753	2.750	2.742	2.737
occupancy x	0.506(1)	0.504(1)	0.498(1)	0.4997(9)	0.4897(8)
F(000) ^a	554.1	554.1	554.1	554.1	554.1
abs. coeff. (mm ⁻¹)	10.53	10.51	10.53	10.49	10.54
2 θ range (deg)	3.2-80.50	3.2-78.7	3.2-75.6	3.2-84.2	3.2-75.5
index range h, k, l	-16 16, -22 22, -11 11	-16 16, -21 21, -11 11	-15 15, -21 21, -10 10	-17 17, -23 22, -11 11	-15 15, -21 21, -10 10
no. refl. collected	8518	8138	7459	8732	7453
no. unique refl.	4346	4121	3744	4838	3758
R equivalent refl.	0.022	0.019	0.021	0.029	0.024
data completeness	0.988	0.987	0.998	0.981	0.999
GOF ^b on F	1.29	1.21	1.32	1.18	1.27
no. refl. > 5 σ (I)	3031	2579	2535	2595	2199
R ₁ [I > 5 σ (I)] ^c	0.019	0.019	0.018	0.020	0.018
wR ₂ [I > 5 σ (I)] ^d	0.020	0.020	0.019	0.023	0.019
extinct. coeff.	6.920×10^{-7}	5.405×10^{-7}	6.968×10^{-7}	8.617×10^{-7}	9.268×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.51/-0.56	0.40/-0.62	0.40/-0.43	0.33/-0.36	0.35/-0.41

Table S-H. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.498$

	T = 270 K	T = 298 K
formula	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$	$CuH_{12}K_{0.996}O_{14}Rb_{1.004}Se_2$
fw ^a	582.31	582.31
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$
a (Å)	9.2572(3)	9.2654(2)
b (Å)	12.3887(4)	12.4017(4)
c (Å)	6.3667(2)	6.3686(2)
β° (deg)	104.062(1)	104.114(2)
V (Å ³)	708.28(4)	709.70(3)
Z	2	2
d_{calc} (g cm ⁻³) ^a	2.730	2.725
occupancy x	0.497(1)	0.4894(8)
F(000) ^a	554.1	554.1
abs. coeff. (mm ⁻¹)	10.47	10.49
2 θ range (deg)	3.2-84.0	3.2-78.7
index range h, k, l	-17 17, -23 22, -11 11	-16 16, -21 21, -11 11
no. refl. collected	9599	7683
no. unique refl.	4874	4074
R equivalent refl.	0.027	0.025
data completeness	0.989	0.965
GOF ^b on F	1.33	1.36
no. refl. > 5 σ (I)	2436	2053
R_1 [$I > 5\sigma(I)$] ^c	0.019	0.019
wR_2 [$I > 5\sigma(I)$] ^d	0.021	0.020
extinct. coeff.	8.710×10^{-7}	8.754×10^{-7}
largest diff. peak and hole (e Å ⁻³)	0.41/-0.36	0.40/-0.31

Table S-I. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.728$

	T = 90 K	T = 140 K	T = 190 K	T = 220 K	T = 250 K
formula	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$
fw ^a	560.98	560.98	560.98	560.98	560.98
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.1628(2)	9.1728(2)	9.1885(2)	9.1940(2)	9.2045(2)
b (Å)	12.2679(3)	12.2762(3)	12.2942(3)	12.3017(3)	12.3143(4)
c (Å)	6.3358(2)	6.3399(2)	6.3469(1)	6.3483(2)	6.3515(2)
β° (deg)	103.367(1)	103.437(1)	103.517(1)	103.592(1)	103.654(2)
V (Å ³)	692.90(3)	694.38(3)	697.12(2)	697.90(3)	699.58(3)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.689	2.683	2.672	2.669	2.663
occupancy x	0.728(1)	0.7325(6)	0.7293(7)	0.7254(8)	0.7229(9)
F(000) ^a	537.6	537.6	537.6	537.6	537.6
abs. coeff. (mm ⁻¹)	9.23	9.18	9.16	9.18	9.17
2 θ range (deg)	3.2-84.2	3.2-84.2	3.2-80.5	3.2-80.5	3.2-78.8
index range h, k, l	-17 16, -22 23, -11 11	-17 16, -22 22, -11 11	-16 16, -21 22, -11 11	-16 16, -21 22, -11 11	-16 16, -21 21, -11 11
no. refl. collected	9432	9280	8412	8333	7674
no. unique refl.	4782	4777	4302	4326	4094
R equivalent refl.	0.024	0.023	0.021	0.020	0.020
data completeness	0.985	0.981	0.982	0.986	0.981
GOF ^b on F	1.34	1.28	1.37	1.40	1.39
no. refl. > 5 σ (I)	3257	3166	2854	2670	2554
R ₁ [$I > 5\sigma(I)$] ^c	0.021	0.020	0.020	0.020	0.021
wR ₂ [$I > 5\sigma(I)$] ^d	0.022	0.020	0.020	0.021	0.021
extinct. coeff.	3.539×10^{-6}	3.109×10^{-6}	3.544×10^{-6}	2.898×10^{-6}	3.101×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.77/-0.73	0.50/-0.50	0.48/-0.53	0.49/-0.51	0.51/-0.42

Table S-I. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 0.728$

	T = 298 K
formula	$CuH_{12}K_{1.456}O_{14}Rb_{0.544}Se_2$
fw ^a	560.98
wavelength (Å)	0.71069
crystal system	monoclinic
space group	$P2_1/a$
a (Å)	9.2207(2)
b (Å)	12.3347(3)
c (Å)	6.3561(2)
β° (deg)	103.767(2)
V (Å ³)	702.14(3)
Z	2
d_{calc} (g cm ⁻³) ^a	2.653
occupancy x	0.7282(8)
F(000) ^a	537.6
abs. coeff. (mm ⁻¹)	9.10
2 θ range (deg)	3.2-75.5
index range h, k, l	-15 15, -21 20, -10 10
no. refl. collected	7297
no. unique refl.	3715
R equivalent refl.	0.023
data completeness	0.992
GOF ^b on F	1.34
no. refl. > 5 σ (I)	2287
R_1 [$I > 5\sigma(I)$] ^c	0.018
wR_2 [$I > 5\sigma(I)$] ^d	0.020
extinct. coeff.	3.106×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.37/-0.34

Table S-J. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 1.000$

	T = 85 K	T = 170 K	T = 210 K	T = 240 K	T = 298 K
formula	$CuH_{12}K_2O_{14}Se_2$	$CuH_{12}K_2O_{14}Se_2$	$CuH_{12}K_2O_{14}Se_2$	$CuH_{12}K_2O_{14}Se_2$	$CuH_{12}K_2O_{14}Se_2$
fw ^a	535.75	535.75	535.75	535.75	535.75
wavelength (Å)	0.71069	0.71069	0.71069	0.71069	0.70169
crystal system	monoclinic	monoclinic	monoclinic	monoclinic	monoclinic
space group	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$	$P2_1/a$
a (Å)	9.1216(1)	9.1392(1)	9.1496(2)	9.1617(2)	9.1771(1)
b (Å)	12.2177(2)	12.2346(2)	12.2443(3)	12.2559(2)	12.2753(2)
c (Å)	6.3082(1)	6.3211(1)	6.3270(1)	6.3327(1)	6.3405(1)
β° (deg)	102.9339(7)	103.0477(7)	103.154(1)	103.1828(8)	103.3209(8)
V (Å ³)	685.18(2)	688.54(2)	690.22(2)	692.33(3)	695.05(2)
Z	2	2	2	2	2
d_{calc} (g cm ⁻³) ^a	2.597	2.584	2.578	2.570	2.560
occupancy x	1.000	1.000	1.000	1.000	1.000
F(000) ^a	518.0	518.0	518.0	518.0	518.0
abs. coeff. (mm ⁻¹)	7.58	7.54	7.52	7.50	7.47
2 θ range (deg)	3.2-84.2	3.2-78.7	3.2-69.9	3.2-68.7	3.2-80.6
index range h, k, l	-17 17, -22 23, -11 11	-16 16, -20 21, -11 11	-14 14, -16 19, -10 10	-14 14, -10 10, -15 19	-16 16, -21 22, -11 11
no. refl. collected	9254	7974	5382	5066	8358
no. unique refl.	4809	4086	3020	2878	4361
R equivalent refl.	0.019	0.017	0.015	0.016	0.022
data completeness	0.999	0.997	0.998	0.998	0.994
GOF ^b on F	1.44	1.50	1.44	1.39	1.39
no. refl. > 5 σ (I)	3828	3306	2378	2212	2896
R ₁ [$I > 5\sigma(I)$] ^c	0.021	0.020	0.018	0.019	0.021
wR ₂ [$I > 5\sigma(I)$] ^d	0.025	0.024	0.021	0.022	0.023
extinct. coeff.	2.419×10^{-6}	2.578×10^{-6}	3.583×10^{-6}	3.517×10^{-6}	2.950×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.64/-1.07	0.47/-0.58	0.60/-0.53	0.63/-0.44	0.47/-0.59

Table S-J. Crystal and Structure Refinement Data for $K_{2x}Rb_{2-2x}[Cu(H_2O)_6](SeO_4)_2$; $x = 1.000$

	T = 320 K
formula	$CuH_{12}K_2O_{14}Se_2$
fw ^a	535.75
wavelength (Å)	0.71069
crystal system	Monoclinic
space group	$P2_1/a$
a (Å)	9.1826(9)
b (Å)	12.2882(9)
c (Å)	6.3449(5)
β° (deg)	103.368(4)
V (Å ³)	696.55(8)
Z	2
d_{calc} (g cm ⁻³) ^a	2.554
occupancy x	1.000
F(000) ^a	518.0
abs. coeff. (mm ⁻¹)	7.46
2 θ range (deg)	3.2-75.6
index range h, k, l	-10 10, -21 17, -10 10
no. refl. collected	4736
no. unique refl.	2869
R equivalent refl.	0.020
data completeness	0.771
GOF ^b on F	1.29
no. refl. > 5 σ (I)	1991
R_1 [$I > 5\sigma(I)$] ^c	0.021
wR_2 [$I > 5\sigma(I)$] ^d	0.027
extinct. coeff.	2.221×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.36/-0.41

Table S-K. Crystal and Structure Refinement Data for $\text{Rb}_2[\text{Zn}(\text{H}_2\text{O})_6](\text{SeO}_4)_2$ and $\text{K}_2[\text{Zn}(\text{H}_2\text{O})_6](\text{SeO}_4)_2$

	T = 100 K	T = 298 K
formula	$\text{H}_{12}\text{O}_{14}\text{Rb}_2\text{Se}_2\text{Zn}$	$\text{H}_{12}\text{K}_2\text{O}_{14}\text{Se}_2\text{Zn}$
fw ^a	630.33	537.59
wavelength (Å)	0.71069	0.71069
crystal system	monoclinic	Monoclinic
space group	$\text{P}2_1/\text{a}$	$\text{P}2_1/\text{a}$
a (Å)	9.3149(2)	9.2140(2)
b (Å)	12.5466(3)	12.3957(3)
c (Å)	6.3433(1)	6.2785(1)
β° (deg)	105.025(1)	104.201(1)
V (Å ³)	716.00(2)	695.18(2)
Z	2	2
d_{calc} (g cm ⁻³) ^a	2.923	2.568
occupancy x	0.000	1.000
F(000) ^a	592.0	520.0
abs. coeff. (mm ⁻¹)	13.60	7.67
2 θ range (deg)	3.2-71.3	3.2-75.6
index range h, k, l	-15 15, -20 18, -10 10	-15 15, -21 20, -10 10
no. refl. collected	6265	7048
no. unique refl.	3278	3718
R equivalent refl.	0.023	0.018
data completeness	0.987	0.999
GOF ^b on F	1.42	1.43
no. refl. > 5 σ (I)	2507	2570
R_1 [$I > 5\sigma(I)$] ^c	0.019	0.017
wR_2 [$I > 5\sigma(I)$] ^d	0.021	0.019
extinct. coeff.	3.795×10^{-6}	2.744×10^{-6}
largest diff. peak and hole (e Å ⁻³)	0.76/-0.67	0.42/-0.40

^aThe formula weight, density and number of electrons in the unit cell have been computed on the basis of the value of the occupancy (x) averaged over all the data sets for each crystal. ^bGOF = goodness-of-fit = $[\sum(|F_o| - |F_c|)^2 / (m - p)]^{1/2}$ where m is the number of reflections and p the number of variables. ^c $R_1 = \sum||F_o| - |F_c|| / \sum|F_o|$. ^d $wR_2 = [\sum w(|F_o| - |F_c|)^2 / \sum w|F_o|^2]^{1/2}$; $w = 1/\sigma^2(F_o)$.