Supporting Information

for

Pressure-Induced Metathesis Reaction to Sequester Cs

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Supporting Figure

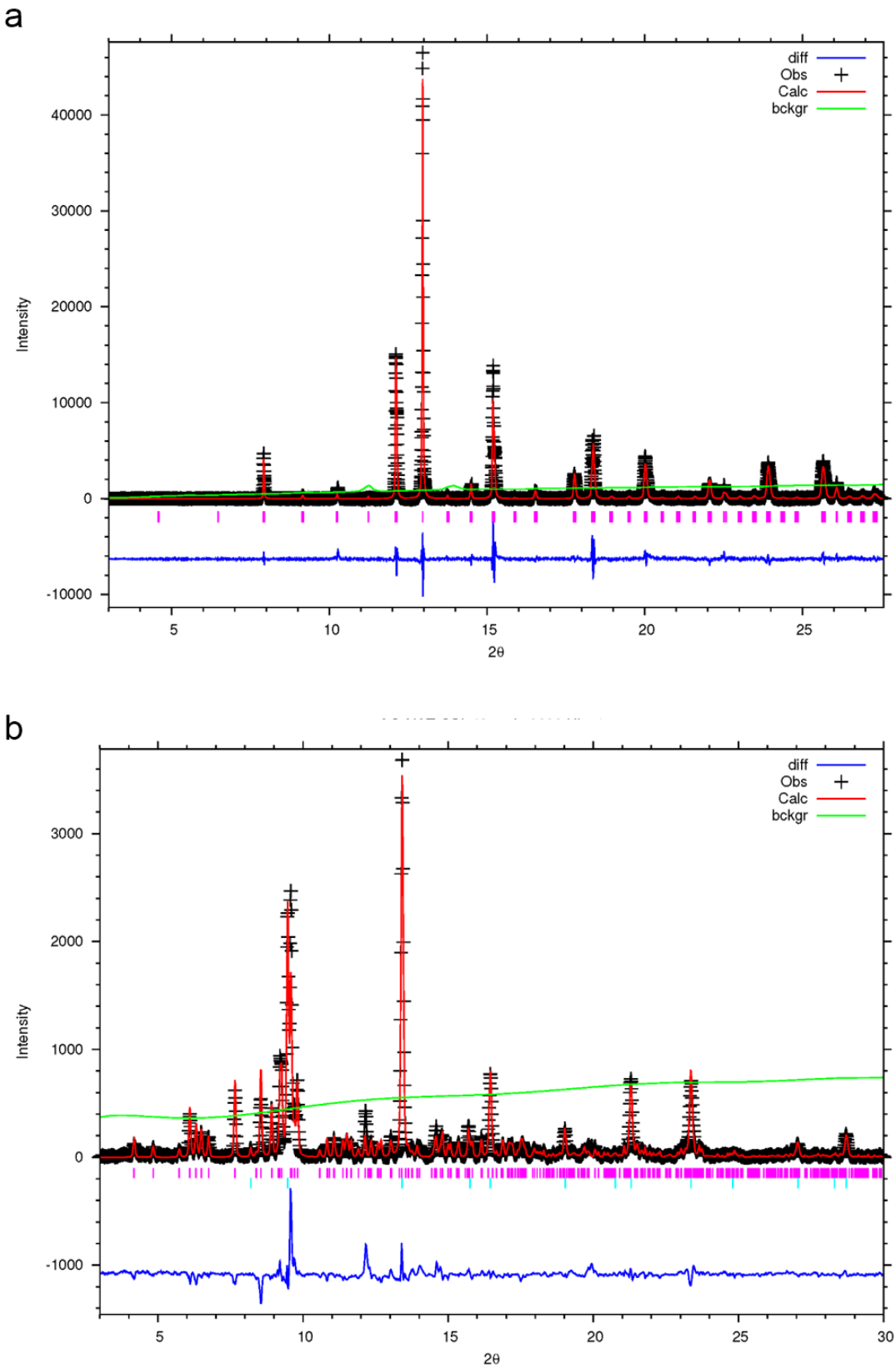

Figure S1. Representative final Rietveld refinement fits. (a) monoclinic pollucite after pressure release in dehy-Cs-NAT in water PTM measured at the X14A beamline at the NSLS (λ=0.77486 Å). (b) Ag-NAT at 1.42 GPa in CsI 1M PTM measured at the 10C beamline at the PAL (λ=0.4959 Å).

Supporting Tables

Table S1. Changes of the unit-cell parameters as a function of pressure and heat treatment. [a]

	P (GPa)	treatment	phase	space group	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)	V (Å^3)	V (O_{80}) (Å^3)
Ag-NAT to pollucite	0.33		Ag-NAT	**F***dd2*	18.553(3)	18.915(2)	6.5579(6)				2301.4(5)	
			Cs-NAT-I	**F***dd2*	19.925(2)	20.312(3)	6.5454(5)				2655.9(6)	
	0.55		Cs-NAT-II	**F***dd2*	18.770(1)	20.175(2)	6.5063(4)				2463.9(4)	
	0.98		Cs-NAT-II	**F***dd2*	18.656(1)	19.924(1)	6.4970(3)				2415.0(2)	
	0.98	130 ℃ heated	Cs-NAT-II	**F***dd2*	18.655(1)	19.883(1)	6.4898(4)				2407.3(4)	
	1.42		Cs-NAT-II	**F***dd2*	18.579(1)	19.756(1)	6.4835(4)				2379.7(3)	
	1.42	140 ℃ heated	Cs-NAT-II	**F***dd2*	18.601(2)	19.799(2)	6.4677(5)				2381.9(5)	
	2.07		Cs-NAT-II	**F***dd2*	18.466(2)	19.574(1)	6.4703(4)				2338.8(4)	
	2.07	160 ℃ heated	Cs-NAT-II	**F***dd2*	18.519(1)	19.626(1)	6.4750(2)				2353.3(1)	
			triclinic-pollucite	**P**$\bar{1}$	13.3509(6)	13.4052(4)	13.3468(6)	91.277(3)	88.953(3)	89.134(4)	2387.4(1)	1989.5(1)
	2.61		triclinic-pollucite	**P**$\bar{1}$	13.3132(5)	13.3311(5)	13.2873(4)	91.550(4)	89.022(2)	88.387(4)	2356.0(2)	1963.4(2)
	2.61	165 ℃ heated	triclinic-pollucite	**P**$\bar{1}$	13.2981(8)	13.3329(6)	13.2932(7)	91.268(5)	88.796(4)	88.484(4)	2355.0(2)	1962.5(2)
	2.94		triclinic-pollucite	**P**$\bar{1}$	13.2570(4)	13.2640(5)	13.2464(5)	91.598(3)	88.897(3)	88.756(3)	2327.4(2)	1939.4(2)
	0	released	cubic-pollucite[b]	**I***a*$\bar{3}$*d*	13.6240(9)						2528.8(5)	2107.3(5)
Cs-NAT-I to pollucite	0	ambient	Cs-NAT-I	**F***dd2*	20.0078	20.3209	6.56526				2669.286	
	0.52		Cs-NAT-I	**F***dd2*	19.9338	20.3286	6.56276				2659.403	
	1		Cs-NAT-II	**F***dd2*	18.7135(3)	20.0744(4)	6.5209(1)				2449.7(1)	
	1	125 ℃ heated	Cs-NAT-II	**F***dd2*	18.690(1)	19.997(1)	6.5249(4)				2438.6(4)	
	1.64		Cs-NAT-II	**F***dd2*	18.6092(5)	19.8718(5)	6.5087(2)				2406.9(2)	
	1.64	150 ℃ heated	Cs-NAT-II	**F***dd2*	18.6443(7)	19.9196(8)	6.5206(2)				2421.6(2)	
	2.29		Cs-NAT-II	**F***dd2*	18.563(1)	19.763(2)	6.5118(5)				2388.9(5)	
	2.29	160 ℃ heated	triclinic-pollucite	**P**$\bar{1}$	13.3759(8)	13.3861(7)	13.380(1)	90.514(4)	89.083(3)	88.785(3)	2394.7(4)	1995.6(4)

	0	ambient	dehy-Cs-NAT	**F***dd2*	17.9956(9)	18.9323(9)	6.5617(2)				2235.6(3)	
	0.52		dehy-Cs-NAT	**F***dd2*	17.8533(9)	18.8219(9)	6.5396(3)				2197.5(3)	
	0.52	100 ℃ heated	Cs-NAT-II	**F***dd2*	18.8547(6)	20.3042(6)	6.5381(2)				2503.0(2)	
	0.83		Cs-NAT-II	**F***dd2*	18.7889(8)	20.1721(8)	6.5315(2)				2475.5(3)	
	0.83	120 ℃ heated	Cs-NAT-II	**F***dd2*	18.7840(6)	20.1601(7)	6.5314(2)				2473.3(2)	
	1.3		Cs-NAT-II	**F***dd2*	18.6826(5)	19.9698(6)	6.5205(2)				2432.7(2)	
	1.3	140 ℃ heated	Cs-NAT-II	**F***dd2*	18.6977(6)	19.9927(6)	6.5217(2)				2437.9(2)	
	1.5		Cs-NAT-II	**F***dd2*	18.6413(6)	19.8935(6)	6.5159(2)				2416.4(2)	
dehy-Cs-NAT to pollucite	1.5	145 ℃ heated	Cs-NAT-II	**F***dd2*	18.6288(4)	19.8814(5)	6.5164(1)				2413.4(1)	
			triclinic-pollucite	$\mathbf{P}\bar{1}$	13.474(1)	13.4839(6)	13.473(1)	90.712(5)	89.703(8)	88.986(5)	2447.2(2)	2039.4(2)
	2.03		Cs-NAT-II	**F***dd2*	18.5729(5)	19.7943(5)	6.5109(1)				2393.6(2)	
			triclinic-pollucite	$\mathbf{P}\bar{1}$	13.438(1)	13.4428(8)	13.4120(7)	90.876(5)	89.944(5)	88.600(5)	2421.9(2)	2018.2(2)
	2.03	160 ℃ heated	triclinic-pollucite	$\mathbf{P}\bar{1}$	13.4021(5)	13.4060(5)	13.4013(6)	90.893(3)	89.193(2)	88.943(2)	2406.4(2)	2005.4(2)
	2.55		triclinic-pollucite	$\mathbf{P}\bar{1}$	13.3651(4)	13.3798(5)	13.3776(5)	90.504(3)	88.854(1)	88.359(2)	2390.7(2)	1992.2(2)
	2.55	165 ℃ heated	triclinic-pollucite	$\mathbf{P}\bar{1}$	13.3414(6)	13.4190(5)	13.3391(6)	90.973(2)	88.624(2)	88.320(2)	2386.0(3)	1988.3(2)
	3.69		triclinic-pollucite	$\mathbf{P}\bar{1}$	13.2312(4)	13.2980(4)	13.2598(5)	91.091(2)	88.932(2)	87.956(2)	2330.7(2)	1942.3(2)
	0	released	monoclinic-pollucite	**C**2/c	13.6707(4)	19.2945(2)	11.8611(3)		125.208(1)		2556.3(1)	2130.3(2)

Ideal unit-cell composition of Cs-NAT-I : $Cs_{16}Al_{16}Si_{24}O_{80}\cdot 16H_2O$

Ideal Unit-cell composition of dehy-Cs-NAT : $Cs_{16}Al_{16}Si_{24}O_{80}$

Ideal Unit-cell composition of Ag-NAT : $Ag_{16}Al_{16}Si_{24}O_{80}\cdot 16H_2O$

Ideal Unit-cell composition of Cs-NAT-II : $Cs_{16}Al_{16}Si_{24}O_{80}\cdot 16H_2O$

Ideal Unit-cell composition of pollucite : $Cs_{16}Al_{16}Si_{32}O_{96}$

[a]ESD's are in parentheses. All framework sites are fully occupied.

[b]XRD patterns of Ag-NAT to pollucite was measured at beamline 10C at PAL using imaging plate detector. Due to the low resolution in XRD pattern, the recovered pollucite model was refined in cubic.

Table S2. Refined atomic coordinates for dehy-Cs-NAT to pollucite and Ag-NAT to pollucite as a function of pressure and heat treatment.[a,e]

		dehy-Cs-NAT to Pollucite										Ag-NAT to Pollucite
		0.00 GPa	0.52 GPa		0.83 GPa		1.30 GPa		1.50 GPa		2.03 GPa	1.42 GPa
treatment		ambient		100℃		120℃		140℃		145℃		
Space group		**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*	**F***dd2*
$_wR_p$(%), χ^2		7.96, 1.452	6.15, 4.153	4.71, 2.286	7.06, 4.208	6.27, 2.479	5.54, 3.871	6.56, 5.491	6.46, 5.084	4.80, 3.008	4.44, 2.326	5.05, 1.862
Cell parameters	*a*	17.9956(9)	17.8533(9)	18.8547(6)	18.7889(8)	18.7840(6)	18.6826(5)	18.6977(6)	18.6413(6)	18.6288(4)	18.5729(5)	18.580(3)
	b	18.9323(9)	18.8219(9)	20.3042(6)	20.1721(8)	20.1601(7)	19.9698(6)	19.9927(6)	19.8935(6)	19.8814(5)	19.7943(5)	19.750(3)
	c	6.5617(2)	6.5396(3)	6.5381(2)	6.5315(2)	6.5314(2)	6.5205(2)	6.5217(2)	6.5159(2)	6.5164(1)	6.5109(1)	6.4894(8)
Cell volume	*V*	2235.6(3)	2197.5(3)	2503.0(2)	2475.5(3)	2473.3(2)	2432.7(2)	2437.9(2)	2416.4(2)	2413.4(1)	2393.6(2)	2381.3(7)
Si(1) 8a	*x*	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
	y	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
	z	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
	U_{iso}[b]	0.036(1)	0.0472(9)	0.016[c]	0.016(1)	0.016(1)	0.0301(9)	0.019(1)	0.0168(9)	0.020(1)	0.019(1)	0.0089[d]
Si(2) 16b	*x*	0.1495(3)	0.1530(4)	0.1595(3)	0.1579(4)	0.1588(3)	0.1577(3)	0.1576(4)	0.1571(4)	0.1571(3)	0.1553(3)	0.1581(4)
	y	0.2150(3)	0.2142(3)	0.2034(2)	0.2053(3)	0.2053(3)	0.2051(3)	0.2048(4)	0.2055(3)	0.2070(3)	0.2101(3)	0.2057(4)
	z	0.6190(5)	0.6225(9)	0.6188(6)	0.6201(9)	0.6198(7)	0.6188(8)	0.619(1)	0.619(1)	0.6204(8)	0.6212(7)	0.6178(7)
Al(1) 16b	*x*	0.0396(3)	0.0393(4)	0.0518(3)	0.0506(4)	0.0516(3)	0.0498(3)	0.0497(4)	0.0496(4)	0.0497(3)	0.0463(3)	0.0485(4)
	y	0.0936(2)	0.0944(3)	0.0812(2)	0.0827(3)	0.0818(3)	0.0831(3)	0.0834(4)	0.0838(4)	0.0836(3)	0.0877(3)	0.0844(4)
	z	0.6101(5)	0.6141(9)	0.6088(6)	0.6093(9)	0.6093(7)	0.6109(8)	0.612(1)	0.611(1)	0.6100(8)	0.6100(7)	0.6104(7)
O(1) 16b	*x*	0.0334(5)	0.0361(6)	0.0415(4)	0.0390(6)	0.0385(6)	0.0402(5)	0.0391(6)	0.0408(6)	0.0415(6)	0.0334(7)	0.0481(7)
	y	0.0631(3)	0.0630(4)	0.0535(3)	0.0558(4)	0.0563(4)	0.0564(4)	0.0573(4)	0.0563(5)	0.0557(4)	0.0605(4)	0.0505(6)
	z	0.8610(6)	0.864(1)	0.8605(7)	0.862(1)	0.8624(8)	0.8644(9)	0.866(1)	0.864(1)	0.8632(9)	0.8628(8)	0.8595(7)
	U_{iso}[b]	0.036(1)	0.0472(9)	0.016[c]	0.016(1)	0.016(1)	0.0301(9)	0.019(1)	0.0168(9)	0.020(1)	0.019(1)	0.011[d]
O(2) 16b	*x*	0.0664(2)	0.0685(3)	0.0873(3)	0.0849(4)	0.0865(3)	0.0830(3)	0.0827(4)	0.0813(4)	0.0825(4)	0.0792(3)	0.0810(5)
	y	0.1824(2)	0.1830(3)	0.1607(3)	0.1633(4)	0.1621(3)	0.1649(3)	0.1651(4)	0.1664(4)	0.1659(3)	0.1704(3)	0.1676(4)
	z	0.608(1)	0.614(2)	0.603(1)	0.605(2)	0.609(1)	0.607(2)	0.605(2)	0.607(2)	0.603(1)	0.609(1)	0.608(1)
O(3) 16b	*x*	0.1034(4)	0.1015(5)	0.1041(4)	0.1048(5)	0.1067(4)	0.1045(5)	0.1044(5)	0.1050(5)	0.1069(5)	0.1050(5)	0.1009(5)
	y	0.0420(3)	0.0424(3)	0.0239(3)	0.0265(4)	0.0250(3)	0.0263(3)	0.0250(4)	0.0271(4)	0.0295(3)	0.0322(3)	0.0325(3)
	z	0.475(1)	0.473(2)	0.478(1)	0.479(2)	0.484(2)	0.481(2)	0.488(2)	0.481(2)	0.476(2)	0.485(2)	0.452(2)
O(4) 16b	*x*	0.2015(3)	0.2002(4)	0.2172(3)	0.2162(4)	0.2187(3)	0.2151(4)	0.2154(4)	0.2147(4)	0.2139(4)	0.2123(4)	0.2094(4)
	y	0.1617(4)	0.1574(5)	0.1621(4)	0.1628(5)	0.1625(4)	0.1615(5)	0.1615(5)	0.1619(5)	0.1633(5)	0.1620(4)	0.1636(5)
	z	0.749(1)	0.755(2)	0.750(1)	0.747(2)	0.738(2)	0.747(2)	0.747(2)	0.747(2)	0.752(2)	0.739(2)	0.773(2)
O(5) 16b	*x*	0.1828(3)	0.1824(4)	0.1908(3)	0.1894(4)	0.1881(4)	0.1891(4)	0.1889(4)	0.1892(4)	0.1888(4)	0.1846(4)	0.1956(6)
	y	0.2207(5)	0.2197(5)	0.2143(4)	0.2162(6)	0.2173(5)	0.2148(5)	0.2153(6)	0.2142(6)	0.2153(6)	0.2218(7)	0.2063(7)
	z	0.3905(6)	0.390(0)	0.3909(7)	0.392(1)	0.3898(8)	0.390(1)	0.391(1)	0.390(1)	0.3909(9)	0.3900(8)	0.3927(7)
Cs(1) 16b	*x*	0.0441(2)	0.0436(2)	0.0276(1)	0.0274(1)	0.0277(1)	0.0272(1)	0.0264(1)	0.0263(1)	0.0278(1)	0.0273(2)	0.0228(4)
	y	0.1965(1)	0.1960(1)	0.1877(1)	0.1896(1)	0.1896(1)	0.1919(1)	0.1917(1)	0.1922(1)	0.1931(2)	0.1934(2)	0.1964(4)
	z	0.117(1)	0.125(1)	0.1254(9)	0.130(1)	0.140(1)	0.138(1)	0.138(1)	0.136(1)	0.137(1)	0.137(1)	0.134(3)
	U_{iso}[b]	0.036(1)	0.0472(9)	0.016[c]	0.016(1)	0.016(1)	0.0301(9)	0.019(1)	0.0168(9)	0.020(1)	0.019(1)	0.055(3)
OW(1) 16b	*x*			0.1684(8)	0.168(1)	0.1691(9)	0.1616(8)	0.161(1)	0.160(1)	0.164(1)	0.160(1)	0.154(3)
	y			0.0845(7)	0.083(1)	0.0854(8)	0.0842(8)	0.083(1)	0.082(1)	0.090(1)	0.089(1)	0.098(2)
	z			0.024(2)	0.029(3)	0.044(2)	0.050(2)	0.042(3)	0.039(3)	0.058(3)	0.034(3)	0.081(9)
	U_{iso}[b]			0.016[b]	0.016(1)	0.016(1)	0.0301(9)	0.019(1)	0.0168(9)	0.020(1)	0.019(1)	0.055(3)

[a]ESD's are in parentheses All framework sites are fully occupied.

[b]Isotropic displacement factors (U_{iso}) were refined by grouping.

[c]U_{iso}'s fixed to the values of the next model.

[d]U_{iso}'s fixed to the values of the reference model. (by Y. Lee et al., 2010)

[e]CIFs' show a- and b-class alert due to highly estimated standard deviation measured in T-O-T and O-T-O angles. These are caused from the heavy restraints applied in Si/Al-tetrahedra for high-pressure XRD data. Also, there are no bonds between T-M atoms. These are caused from the imaginary bonding distances in CIF calculation, thus they can be negligible. Lastly, hydrogen atoms are not refined in these refinements and all wavelength and temperature information are included in CIFs.

Table S3. Selected interatomic distances (Å) and angles (°) for dehy-Cs-NAT to pollucite and Ag-NAT to pollucite as a function of pressure and heat treatment.[a,c]

	dehy-Cs-NAT to Pollucite										Ag-NAT to Pollucite
	0.00 GPa	0.52 GPa		0.83 GPa		1.30 GPa		1.50 GPa		2.03 GPa	1.42 GPa
treatment	ambient		100℃		120℃		140℃		145℃		
Si(1) - O(1) x 2	1.6184(8)	1.615(1)	1.619(1)	1.618(1)	1.618(1)	1.617(1)	1.617(2)	1.617(2)	1.618(1)	1.618(1)	1.620(1)
Si(1) - O(5) x 2	1.6188(8)	1.617(1)	1.619(1)	1.619(1)	1.619(1)	1.619(1)	1.620(2)	1.620(2)	1.618(1)	1.618(1)	1.620(1)
mean[b]	1.6186(4)	1.616(1)	1.619(1)	1.619(1)	1.619(1)	1.618(1)	1.619(1)	1.619(1)	1.618(1)	1.618(1)	1.620(1)
Si(2) - O(2)	1.619(1)	1.619(2)	1.616(2)	1.615(2)	1.615(2)	1.612(2)	1.613(2)	1.613(2)	1.616(2)	1.618(2)	1.619(1)
Si(2) - O(3)	1.618(1)	1.614(2)	1.619(2)	1.619(2)	1.617(2)	1.618(2)	1.620(2)	1.619(2)	1.617(2)	1.619(2)	1.619(1)
Si(2) - O(4)	1.618(1)	1.613(2)	1.620(2)	1.618(2)	1.617(2)	1.615(2)	1.615(2)	1.615(2)	1.616(2)	1.617(2)	1.619(1)
Si(2) - O(5)	1.618(1)	1.614(2)	1.618(2)	1.618(2)	1.618(2)	1.616(2)	1.617(2)	1.617(2)	1.616(2)	1.618(2)	1.620(1)
mean[b]	1.618(1)	1.615(1)	1.618(1)	1.618(1)	1.617(1)	1.615(1)	1.616(1)	1.616(1)	1.616(1)	1.618(1)	1.619(1)
Al - O(1)	1.748(1)	1.741(2)	1.750(2)	1.750(2)	1.748(2)	1.746(2)	1.747(2)	1.747(2)	1.748(2)	1.748(2)	1.750(1)
Al - O(2)	1.750(1)	1.748(2)	1.749(2)	1.748(2)	1.747(2)	1.746(2)	1.747(2)	1.747(2)	1.748(2)	1.748(2)	1.750(1)
Al - O(3)	1.747(1)	1.743(2)	1.749(2)	1.748(2)	1.747(2)	1.746(2)	1.748(2)	1.747(2)	1.748(2)	1.749(2)	1.750(1)
Al - O(4)	1.748(1)	1.744(2)	1.751(2)	1.751(2)	1.749(2)	1.749(2)	1.750(2)	1.750(2)	1.749(2)	1.749(2)	1.750(1)
mean[b]	1.748(1)	1.744(1)	1.750(1)	1.749(1)	1.748(1)	1.747(1)	1.748(1)	1.748(1)	1.748(1)	1.749(1)	1.750(1)
Si(1) - O(1) - Al	142.9(3)	141.1(4)	143.0(3)	143.0(4)	142.6(4)	141.1(4)	140.9(5)	141.0(5)	141.2(4)	143.2(4)	139.4(4)
Si(2) - O(2) - Al	128.4(3)	128.6(4)	144.7(4)	143.2(5)	144.7(4)	140.6(4)	139.9(5)	138.5(5)	140.6(4)	139.4(4)	137.9(4)
Si(2) - O(3) - Al	134.5(4)	134.1(5)	135.6(4)	134.8(5)	132.7(4)	135.1(5)	135.9(6)	134.9(6)	131.5(5)	133.9(5)	133.6(5)
Si(2) - O(4) - Al	141.3(4)	135.2(5)	141.9(5)	143.2(6)	143.2(5)	141.9(6)	142.4(7)	142.7(7)	142.5(6)	142.8(5)	138.3(6)
Si(1) - O(5) - Si(2)	145.8(3)	143.0(5)	146.8(4)	147.4(5)	146.0(4)	145.9(5)	146.4(6)	145.6(5)	145.7(4)	145.7(4)	142.0(5)
Average chain rotation angle of T_5O_{10} unit. Ψ	24.9°	24.6°	16.4°	17.4°	16.8°	18.1°	18.2°	18.8°	18.5°	20.1°	19.9°
Cs - O(1)	3.038(7)	3.031(9)	3.239(8)	3.22(1)	3.248(9)	3.251(9)	3.23(1)	3.24(1)	3.27(1)	3.180(9)	3.42(2)
Cs - O(2)	3.262(9)	3.24(1)	3.362(8)	3.33(1)	3.303(9)	3.28(1)	3.27(1)	3.27(1)	3.25(1)	3.25(1)	3.31(2)
	3.036(5)	3.033(6)									3.31(1)
Cs - O(3)	3.511(7)	3.497(9)	3.429(7)	3.38(1)	3.347(8)	3.344(9)	3.35(1)	3.31(1)	3.282(9)	3.298(9)	3.25(1)
Cs - O(4)	3.643(8)	3.569(9)	3.375(8)	3.37(1)	3.262(9)	3.332(9)	3.32(1)	3.34(1)	3.40(1)	3.33(1)	3.53(2)
Cs - O(5)	3.110(6)	3.056(7)	3.50(1)	3.533(8)	3.472(6)	3.472(7)	3.490(8)	3.484(8)	3.454(8)	3.401(7)	3.34(2)
Cs - Cs	4.170(3)	4.154(3)	4.263(3)	4.204(3)	4.205(3)	4.128(3)	4.129(3)	4.107(3)	4.100(3)	4.080(4)	3.966(9)
OW - O(1)			2.70(1)	2.71(2)	2.79(2)	2.63(2)	2.60(2)	2.54(2)	2.70(2)	2.67(2)	2.61(5)
			3.30(2)	3.25(2)	3.16(2)	3.27(2)	3.29(2)	3.34(2)	3.18(2)	3.08(2)	3.35(5)
OW - O(2)			3.38(2)	3.35(2)	3.37(2)	3.44(2)	3.44(2)	3.42(2)	3.42(2)	3.34(2)	3.62(6)
OW - O(3)			3.43(1)	3.36(2)	3.34(2)	3.22(2)	3.31(2)	3.25(2)	3.16(2)	3.30(2)	2.90(6)
OW - O(4)			2.56(1)	2.62(2)	2.70(2)	2.70(2)	2.69(2)	2.68(2)	2.64(2)	2.60(2)	2.60(6)
OW - O(5)			3.28(2)	3.18(2)	3.18(2)	3.09(2)	3.07(2)	3.06(2)	3.18(2)	2.99(3)	3.04(5)
OW - Cs			3.12(2)	3.12(2)	3.09(2)	3.13(2)	3.16(2)	3.18(2)	3.09(2)	3.22(2)	3.09(6)

[a]ESD's are in parentheses

[b]Mean values are computed using $\sigma = \frac{1}{n}\left[\sum_{i=1}^{n}\sigma_i^2\right]^{\frac{1}{2}}$

[e]CIFs' show a- and b-class alerts due to highly estimated standard deviation measured in T-O-T and O-T-O angles. These are caused from the heavy restraints applied in Si/Al-tetrahedra for high-pressure XRD data. Also, there are no bonds between T-M or O-M atoms. These are caused from the imaginary bonding distances in CIF calculation, thus they can be negligible. Lastly, hydrogen atoms are not refined in these refinements and all wavelength and temperature information are included in CIFs.

Table S4. Refined atomic coordinates for the recovered monoclinic pollcuite.[a,c]

Space Group	C 2/c				
Cell parameters	a	b	c	beta	
	13.6710(4)	19.2938(3)	11.8606(3)	125.205(1)	
$_{w}R_{p}$(%), χ^2	8.90 , 0.114				

	x	*y*	*z*	*Uiso*[b]	*Occu.*
Si(1)	0.4003(1)	0.1341(1)	0.4523(1)	0.022(2)	0.6667
Si(2)	0.2317(1)	0.0015(1)	0.2729(1)	0.022(2)	0.6667
Si(3)	0.1913(1)	0.2237(1)	0.2215(1)	0.022(2)	0.6667
Si(4)	0.1689(1)	0.0964(1)	0.0446(1)	0.022(2)	0.6667
Si(5)	0.3340(1)	-0.1388(1)	0.4404(1)	0.022(2)	0.6667
Si(6)	0	-0.0402(2)	0.25	0.022(2)	0.6667
Si(7)	0	0.3072(2)	0.25	0.022(2)	0.6667
Al(1)	0.4003(1)	0.1341(1)	0.4523(1)	0.022(2)	0.3333
Al(2)	0.2317(1)	0.0015(1)	0.2729(1)	0.022(2)	0.3333
Al(3)	0.1913(1)	0.2237(1)	0.2215(1)	0.022(2)	0.3333
Al(4)	0.1689(1)	0.0964(1)	0.0446(1)	0.022(2)	0.3333
Al(5)	0.3340(1)	-0.1388(1)	0.4404(1)	0.022(2)	0.3333
Al(6)	0	-0.0402(2)	0.25	0.022(2)	0.3333
Al(7)	0	0.3072(2)	0.25	0.022(2)	0.3333
O(1)	0.2840(7)	-0.0786(1)	0.3194(4)	0.022(2)	1
O(2)	0.1077(2)	0.0087(2)	0.2671(3)	0.022(2)	1
O(3)	0.3299(5)	0.0592(2)	0.3863(6)	0.022(2)	1
O(4)	0.1038(2)	0.2561(2)	0.2631(3)	0.022(2)	1
O(5)	0.3228(4)	0.2042(2)	0.3655(5)	0.022(2)	1
O(6)	0.1388(4)	0.1515(2)	0.1285(3)	0.022(2)	1
O(7)	0.2039(5)	0.0191(2)	0.1197(3)	0.022(2)	1
O(8)	0.4374(6)	0.1421(2)	0.6119(2)	0.022(2)	1
O(9)	-0.0460(5)	-0.0910(2)	0.1152(2)	0.022(2)	1
O(10)	0.2767(2)	-0.1256(4)	0.5328(3)	0.022(2)	1
O(11)	0.4789(2)	-0.1348(7)	0.5487(4)	0.022(2)	1
O(12)	0.2934(4)	-0.1348(7)	0.5487(4)	0.022(2)	1
Cs(1)	0.6400(4)	0.3747(4)	0.0001(5)	0.0210(5)	0.945(3)
Cs(2)	0	0.4933(5)	0.25	0.0210(5)	0.888(5)
Cs(3)	0.5	0.2429(5)	0.25	0.0210(5)	0.988(5)

[a]ESD's are in parentheses All framework sites are fully occupied.

[b]Isotropic displacement factors (U_{iso}) were refined by grouping.

[c]CIFs' show a-class alert due to highly estimated standard deviation measured in T-O-T and O-T-O angles. These are caused from the heavy restraints applied in Si/Al-tetrahedra for high-pressure XRD data. Also, there are no bonds between T-M or O-M atoms. These are caused from the imaginary bonding distances in CIF calculation, thus they can be negligible.

Table S5. Selected interatomic distances (Å) and angles (°) for the recovered monoclinic pollucite.[a,c]

T1 - O3	1.660(4)		T4 - O6	1.663(5)		T2 - O1 - T5	144.8(5)
T1 - O5	1.660(4)		T4 - O7	1.659(4)		T2 - O2 - T6	140.3(3)
T1 - O8	1.660(4)		T4 - O9	1.659(3)		T1 - O3 - T2	160.3(3)
T1 - O11	1.658(4)		T4 - O10	1.658(4)		T3 - O4 - T7	157.1(2)
mean[b] T1 - O	1.660(2)		mean[b]. T4 - O	1.660(2)		T1 - O5 - T3	138.5(3)
						T3 - O6 - T4	140.6(4)
T2 - O1	1.660(4)		T5 - O1	1.661(4)		T2 - O7 - T4	125.7(3)
T2 - O2	1.662(3)		T5 - O10	1.659(3)		T1 - O8 - T7	145.6(4)
T2 - O3	1.663(4)		T5 - O11	1.658(3)		T4 - O9 - T6	135.2(3)
T2 - O7	1.662(4)		T5 - O12	1.663(4)		T4 - O10 - T5	150.0(2)
mean[b] T2 - O	1.662(2)		mean[b]. T5 - O	1.660(2)		T1 - O11 - T5	140.4(3)
						T3 - O12 - T5	146.5(4)
T3 - O4	1.657(3)		T6 - O2	1.660(3) X2			
T3 - O5	1.658(4)		T6 - O9	1.659(3) X2			
T3 - O6	1.660(4)		mean[b]. T6 - O	1.660(2)			
T3 - O12	1.663(5)						
mean[b] T3 - O	1.660(2)		T7 - O4	1.661(3) X2			
			T7 - O8	1.660(4) X2			
			mean[b]. T7 - O	1.661(2)			
Cs1 - O1	3.230(7)		Cs2 - O7	3.437(7) X2		Cs3 - O9	3.472(9) X2
Cs1 - O2	3.390(8)		Cs2 - O8	2.936(9) X2		Cs3 - O10	3.466(8) X2
Cs1 - O6	3.226(7)		Cs2 - O11	3.53(1) X2		Cs3 - O11	3.06(1) X2
Cs1 - O7	3.018(7)						
Cs1 - O8	3.490(9)						
Cs1 - O9	3.57(1)						

[a]ESD's are in parentheses

[b]Mean values are computed using $$\sigma = \frac{1}{n}\left[\sum_{i=1}^{n}\sigma_i^2\right]^{\frac{1}{2}}$$

[c]CIFs' show a-class alert due to highly estimated standard deviation measured in T-O-T and O-T-O angles. These are caused from the heavy restraints applied in Si/Al-tetrahedra for high-pressure XRD data. Also, there are no bonds between T-M or O-M atoms. These are caused from the imaginary bonding distances in CIF calculation, thus they can be negligible.

Table S6. Bond Valence Sum of the recovered monoclinic pollucite, compared to the hypothetical cubic phase.

	Atom Site	Valence state assumed	Bond Valence Sum
Hypothetical cubic pollucite	Cs	1	0.725999
	Si (Al) / mean	4 (3) / 3.6667	3.8 (3.76)
Monoclinic pollucite	Cs	1	0.8320001
	Si1 (Al1) / mean	4 (3) / 3.6667	3.637
	Si2 (Al2) / mean	4 (3) / 3.6667	3.628
	Si3 (Al3) / mean	4 (3) / 3.6667	3.64
	Si4 (Al4) / mean	4 (3) / 3.6667	3.648
	Si5 (Al5) / mean	4 (3) / 3.6667	3.63
	Si6 (Al6) / mean	4 (3) / 3.6667	3.656
	Si7 (Al7) / mean	4 (3) / 3.6667	3.62