

Table S1. Experimental details of the various DAC high-pressure loadings.

Application parameters	Raman spectra	XRD intensities (crystal structures)	XRD lattice (equation of state)
DAC type	ETH	ETH	ETH
Anvil type and culet C	BA, C=0.6 mm	BA, C=0.6 mm	BA, C=0.6 mm
Thickness of pre-indented gasket	$83 \pm 2 \mu\text{m}$	$90 \pm 2 \mu\text{m}$	$105 \pm 2 \mu\text{m}$
Pressure-chamber diameter	$225 \pm 5 \mu\text{m}$	$260 \pm 5 \mu\text{m}$	$270 \pm 5 \mu\text{m}$
Sample crystal (johnkoivulaite)	$70 \times 70 \times 40 \mu\text{m}^3$	$220 \times 130 \times 40 \mu\text{m}^3$	$150 \times 110 \times 40 \mu\text{m}^3$
XRD pressure calibrant (quartz)	-	-	$90 \times 55 \times 40 \mu\text{m}^3$
Optical pressure sensor	ruby	ruby	ruby
Pressure-transmitting medium ethanol	argon	4:1 methanol-ethanol	4:1 methanol-
Applied pressures	0.0001 to 10.19 GPa	1.65, 6.50 GPa	0.45 to 8.42 GPa

Table S2. Experimental details of the single-crystal X-ray structure investigations (crystal data, intensity data collection, data processing and structure refinements). Statistical parameters as defined by the JANA2006 suite (Petricek et al. 2014).

<i>P</i> (GPa)	0.0001	1.65(6)	6.50(5)
Unit-cell parameters	<i>a</i> = 9.470(1) Å <i>c</i> = 9.050(3) Å <i>V</i> = 702.9(2) Å ³	<i>a</i> = 9.450(1) Å <i>c</i> = 9.010(3) Å <i>V</i> = 696.8(3) Å ³	<i>a</i> = 16.130(2) Å <i>c</i> = 8.850(2) Å <i>V</i> = 1994.1(6) Å ³
Space group	<i>P</i> 6/ <i>mcc</i> (192)	<i>P</i> 6/ <i>mcc</i> (192)	<i>P</i> $\bar{3}$ <i>c</i> 1 (165)
Z [Cs(Be ₂ B)Mg ₂ Si ₆ O ₁₈]	2	2	6
Scans	ω = 0.5°	ω = 0.5°	ω = 0.5°
sin θ /λ	≤ 0.939 Å ⁻¹	≤ 0.799 Å ⁻¹	≤ 0.812 Å ⁻¹
	-17 ≤ <i>h</i> ≤ +17	9 ≤ <i>h</i> ≤ +9	-20 ≤ <i>h</i> ≤ +16
	-17 ≤ <i>k</i> ≤ +10	-5 ≤ <i>k</i> ≤ +6	-20 ≤ <i>k</i> ≤ +24
	-16 ≤ <i>l</i> ≤ +16	-13 ≤ <i>l</i> ≤ +9	-13 ≤ <i>l</i> ≤ +8
Measured reflections	56397	2517	7907
Unique refl. <i>I</i> _o > 0σ(<i>I</i> _o)	859	390	1763
Unique refl. <i>I</i> _o > 3σ(<i>I</i> _o)	785	284	540
Refined parameters	33	30	55
<i>R</i> _{eq}	0.0297	0.065	0.0957
<i>R</i> (<i>F</i>) with <i>I</i> _o > 3σ(<i>I</i> _o)	0.0136	0.0441	0.0560
<i>R</i> (<i>F</i>) with <i>I</i> _o > 0σ(<i>I</i> _o)	0.0157	0.0698	0.2607
<i>wR</i> (<i>F</i>) with <i>I</i> _o > 3σ(<i>I</i> _o)	0.0209	0.0518	0.0448
<i>wR</i> (<i>F</i>) with <i>I</i> _o > 0σ(<i>I</i> _o)	0.0211	0.0528	0.0534
Residuals (e ⁻ /Å ³)	-0.30, +0.65	-1.19, +1.15	-2.59, +2.45

Table S3. Fractional atomic coordinates and atomic displacement parameters (U^{ij} , Å²).

0.0001 GPa	<i>s.o.f</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>Ueq or Uiso</i>	
<i>Cs</i>	0.808(2) <i>Cs</i> , 0.192(2) <i>K</i>	0	0	1/4	0.02399(6)	
<i>Si</i>	1	0.62459(2)	0.72092(2)	1/2	0.00516(6)	
<i>Mg</i>	0.860(4) <i>Mg</i> , 0.140(4) <i>Fe</i>	1/3	2/3	3/4	0.00727(11)	
<i>Be</i>	0.72(7) <i>Be</i> , 0.28(7) <i>B</i>	1/2	1/2	3/4	0.0059(4)	
<i>Ol</i>	1	0.51954(5)	0.64490(5)	0.64777(5)	0.01248(12)	
<i>O2</i>	1	0.78428(7)	0.70078(8)	1/2	0.01390(17)	
	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
<i>Cs</i>	0.02764(7)	0.02764(7)	0.01669(7)	0.01382(4)	0	0
<i>Si</i>	0.00464(8)	0.00434(8)	0.00648(8)	0.00222(6)	0	0
<i>Be</i>	0.0062(5)	0.0062(5)	0.0057(5)	0.0034(4)	0	0
<i>Mg</i>	0.00726(15)	0.00726(15)	0.00728(17)	0.00363(7)	0	0
<i>Ol</i>	0.01336(16)	0.01016(14)	0.01341(14)	0.00549(12)	0.00709(11)	0.00204(11)
<i>O2</i>	0.00865(19)	0.0129(2)	0.0241(3)	0.00827(17)	0	0
1.65 GPa	<i>s.o.f</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>Ueq or Uiso</i>	
<i>Cs</i>	0.818(6) <i>Cs</i> , 0.182(6) <i>K</i>	0	0	1/4	0.0253(5)	
<i>Si</i>	1	0.6243(2)	0.7209(2)	1/2	0.0079(7)	
<i>Mg</i>	0.857(11) <i>Mg</i> , 0.143(11) <i>Fe</i>	1/3	2/3	3/4	0.0096(9)	
<i>Be</i>	0.69(14) <i>Be</i> , 0.31(14) <i>B</i>	1/2	1/2	3/4	0.005(2)	
<i>Ol</i>	1	0.5188(4)	0.6449(4)	0.6482(3)	0.0147(15)	
<i>O2</i>	1	0.7837(6)	0.7008(6)	1/2	0.016(2)	
	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
<i>Cs</i>	0.0294(6)	0.0294(6)	0.0171(7)	0.0147(3)	0	0
<i>Si</i>	0.0076(9)	0.0075(9)	0.0083(9)	0.0035(8)	0	0
<i>Ol</i>	0.0167(18)	0.0144(19)	0.0137(18)	0.0084(15)	0.0031(14)	0.0024(14)
<i>O2</i>	0.013(3)	0.013(3)	0.025(3)	0.010(2)	0	0
6.50 GPa	<i>s.o.f</i>	<i>x</i>	<i>y</i>	<i>z</i>	<i>Ueq or Uiso</i>	
<i>Cs1</i>	0.760(15) <i>Cs</i> , 0.240(15) <i>K</i>	0	0	1/4	0.0202(2)	
<i>Cs2</i>	0.857(8) <i>Cs</i> , 0.143(8) <i>K</i>	2/3	1/3	0.2464(2)	0.0202(2)	
<i>Si1</i>	1	0.1797(4)	0.4509(4)	0.5054(5)	0.0057(3)	
<i>Si2</i>	1	0.1627(3)	0.2207(4)	0.5003(5)	0.0057(3)	
<i>Si3</i>	1	0.3942(4)	0.5092(4)	0.5076(5)	0.0057(3)	
<i>Be1</i>	2/3 <i>Be</i> , 1/3 <i>B</i>	0.1673(13)	0.3395(13)	0.7617(12)	0.0052(3)	
<i>Be2</i>	2/3 <i>Be</i> , 1/3 <i>B</i>	0.498(2)	0	3/4	0.0052(3)	
<i>Mg1</i>	0.975(16) <i>Mg</i> , 0.025(16) <i>Fe</i>	0	0.3324(7)	3/4	0.0086(5)	
<i>Mg2</i>	0.758(15) <i>Mg</i> , 0.242(15) <i>Fe</i>	0.3301(5)	0.3301(5)	3/4	0.0086(5)	
<i>Ol a</i>	1	0.1302(8)	0.3901(8)	0.6563(10)	0.0078(5)	
<i>Ol b</i>	1	0.1240(7)	0.3696(8)	0.3676(11)	0.0078(5)	
<i>Ol c</i>	1	0.1889(7)	0.2706(8)	0.6595(9)	0.0078(5)	
<i>Ol d</i>	1	0.2106(7)	0.2803(7)	0.3543(11)	0.0078(5)	
<i>Ol e</i>	1	0.4553(7)	0.0429(8)	0.6227(11)	0.0078(5)	
<i>Ol f</i>	1	0.4778(6)	0.0767(6)	0.3212(8)	0.0078(5)	
<i>O2 a</i>	1	0.2895(9)	0.4944(8)	0.4687(7)	0.0078(5)	
<i>O2 b</i>	1	0.4543(7)	0.6218(8)	0.5028(11)	0.0078(5)	
<i>O2 c</i>	1	0.1769(7)	0.1317(8)	0.4925(10)	0.0078(5)	
	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
<i>Cs1</i>	0.0228(3)	0.0228(3)	0.0149(3)	0.01139(14)	0	0
<i>Cs2</i>	0.0228(3)	0.0228(3)	0.0149(3)	0.01139(14)	0	0

Table S4. Selected cation-oxygen bond distances (Å).

<i>P</i> (GPa)(Cs,K)	(Be,B)		Si		(Mg,Fe)			
0.0001 GPa (<i>P6/mcc</i>)	<i>Cs-O2</i>	3.396(2) 12×	<i>Be-O1</i>	1.5872(6) 4×	<i>Si-O1</i>	1.6063(6) 2×	<i>Mg-O1</i>	2.0909(7) 6×
					<i>Si-O2</i>	1.6161(9)		
					<i>Si-O2'</i>	1.6170(11)		
1.65(5) GPa (<i>P6/mcc</i>)	<i>Cs-O2</i>	3.386(4) 12×	<i>Be-O1</i>	1.583(4) 4×	<i>Si-O1</i>	1.605(3) 2×	<i>Mg-O1</i>	2.078(5) 6×
					<i>Si-O2</i>	1.610(7)		
					<i>Si-O2'</i>	1.619(7)		
6.50(5) GPa 2.003(11) 2×	<i>Cs1-O2c</i>	3.346(11) 6×	<i>Be1-O1a</i>	1.54(3)	<i>Si1-O1a</i>	1.613(10)	<i>Mg1-O1a</i>	
(<i>P3̄c1</i>)	<i>Cs1-O2c'</i>	3.433(11) 6×	<i>Be1-O1b</i>	1.45(2)	<i>Si1-O1b</i>	1.685(11)	<i>Mg1-O1d</i>	
1.940(15) 2×	<i>Cs2-O2a</i>	3.143(12) 3×	<i>Be1-O1c</i>	1.60(2)	<i>Si1-O2a</i>	1.578(14)	<i>Mg1-O1e</i>	
2.075(16) 2×	<i>Cs2-O2a'</i>	3.552(12) 3×	<i>Be1-O1d</i>	1.60(2)	<i>Si1-O2b</i>	1.636(17)	<i>Mg2-O1b</i>	
2.054(19) 2×	<i>Cs2-O2b</i>	3.256(13) 3×	<i>Be2-O1e</i>	1.64(2) 2×	<i>Si2-O1c</i>	1.572(10)	<i>Mg2-O1c</i>	
2.136(12) 2×	<i>Cs2-O2b'</i>	3.266(13) 3×	<i>Be2-O1f</i>	1.60(2) 2×	<i>Si2-O1d</i>	1.565(10)	<i>Mg2-O1f</i>	2.158(11)
2×					<i>Si2-O2c</i>	1.564(16)		
					<i>Si2-O2c'</i>	1.661(17)		
					<i>Si3-O1e</i>	1.558(15)		
					<i>Si3-O1f</i>	1.620(10)		
					<i>Si3-O2a</i>	1.620(16)		
					<i>Si3-O2b</i>	1.575(12)		

Table S5. Unit-cell parameters, c/a ratio, unit-cell volumes, and unit-cell volume per formula unit (V/Z) of johnkoivulaite, in addition to the unit-cell volume of the quartz used as pressure calibrant under static hydrostatic pressures at 298 K.

V_{Qz} (Å ³)	P (GPa) a (Å)	c (Å)	c/a ' *	V (Å ³)	V/Z (Å ³)	
111.634(4)	0.457(3)	9.4654(8)	9.0323(8)	0.9542	700.83(13)	350.41(7)
110.037(9)	1.034(8)	9.4502(5)	9.0214(4)	0.9546	697.73(8)	348.87(4)
108.188(5)	1.839(4)	9.4349(6)	9.0049(4)	0.9544	694.20(9)	347.10(5)
107.574(4)	2.118(4)	9.4298(7)	8.9983(5)	0.9542	692.94(10)	346.47(5)
106.626(8)	2.570(8)	9.4207(4)	8.9879(3)	0.9541	690.81(6)	345.40(3)
105.578(5)	3.100(5)	9.4091(5)	8.9767(4)	0.9540	688.25(7)	344.14(3)
104.732(10)	3.553(11)	9.4001(5)	8.9657(3)	0.9538	686.09(7)	343.05(3)
103.765(9)	4.098(9)	16.2622(6)	8.9515(5)	0.9534	2050.17(18)	341.70(3)
102.643(6)	4.769(7)	16.2290(5)	8.9285(4)	0.9529	2036.52(15)	339.42(3)
101.783(4)	5.313(4)	16.2003(8)	8.9067(6)	0.9523	2024.39(24)	337.40(4)
100.847(8)	5.935(8)	16.1721(14)	8.8844(5)	0.9515	2012.28(22)	335.38(4)
99.955(11)	6.558(26)	16.1403(5)	8.8610(4)	0.9509	1999.11(16)	333.19(3)
99.350(9)	6.998(21)	16.1200(7)	8.8462(5)	0.9505	1990.75(19)	331.79(3)
n.d.* *	7.50(7)	16.0972(10)	8.8306(7)	0.9502	1981.62(30)	330.27(5)
n.d.* *	8.05(6)	16.0742(7)	8.8140(6)	0.9498	1972.26(21)	328.71(3)
n.d.* *	8.41(6)	16.0534(40)	8.7998(46)	0.9493	1963.98(1.32)	327.33(22)

* $a' = a$ for $P \leq 3.553$ GPa, $a' = a/\sqrt{3}$ for $P \geq 4.098$ GPa

** n.d. = not determined; pressure was determined by the ruby-fluorescence method.