Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.

SUPPLEMENTARY INFORMATION

The crystal structures and mechanical properties of the uranyl carbonate minerals roubaultite, fontanite, sharpite, widenmannite, grimselite and čejkaite

Francisco Colmenero,[a] Jakub Plášil,[b] Jiří Sejkora[c]

[a]*Instituto de Estructura de la Materia (IEM-CSIC). C/ Serrano, 113. 28006 – Madrid, Spain.*

[b]*Institute of Physics ASCR, v.v.i., Na Slovance 2, 182 21, Praha 8, Czech Republic.*

[c]*Mineralogicko-petrologické oddělení, Národní muzeum, Cirkusová 1740, 193 00 Praha 9, Czech Republic.*

Contents: Page

Table S.1. Material data and calculation parameters for the selected uranyl carbonate minerals.

Mineral	Structural formula	Crystal system	Space group	ε(eV)	*k*-mesh
Roubaultite[1]	$Cu_2[(UO_2)_3(CO_3)_2O_2(OH)_2]\cdot 4\,H_2O$	Triclinic	$P\bar{1}\ (no.\ 2)$	1000	2 × 2 × 2
Fontanite[2]	$Ca[(UO_2)_3(CO_3)_2O_2]\cdot 6\,H_2O$	Monoclinic	$P2_1/n\ (no.\ 14)$	950	3 × 1 × 1
Sharpite[3]	$Ca[(UO_2)_3(CO_3)_4]\cdot 3\,H_2O$	Orthorhombic	$Cmcm\ (no.\ 63)$	950	4 × 1 × 1
Widenmannite[4]	$Pb_2[(UO_2)(CO_3)_2(OH)_2]$	Orthorhombic	$Pmmn\ (no.\ 59)$	1000	5 × 3 × 3
Grimselite[5]	$K_3Na[(UO_2)(CO_3)_3]\cdot H_2O$	Hexagonal	$P62c\ (no.\ 190)$	1000	2 × 2 × 2
Čejkaite[6]	$Na_4[(UO_2)(CO_3)_3]$	Monoclinic	$C1c1\ (no.\ 9)$	950	2 × 1 × 4

Table S.2. Interatomic distances in roubaultite (in Å).

Distance	Exp.[1]	Calc.
U1-O		
U1-O1 × 2	1.82(1)	1.844
U1-O4 × 2	2.55(1)	2.535
U1-O5 × 2	2.18(1)	2.185
U1-O6 × 2	2.58(1)	2.627
<U1-Oap>	1.82(1)	1.844
<U1-Oeq>	2.44(1)	2.449
U2-O		
U2-O2	1.81(1)	1.820
U2-O3	1.78(1)	1.806
U2-O4	2.45(1)	2.455
U2-O5	2.25(1)	2.250
U2-O5’	2.25(1)	2.271
U2-O6	2.44(1)	2.429
U2-O8	2.37(1)	2.319
<U2-Oap>	1.80(1)	1.813
<U2-Oeq>	2.35(1)	2.345
Cu-O		
Cu-O7	1.96(1)	2.016
Cu-O7’	2.52(1)	2.448
Cu-O8’	1.95(1)	1.969
Cu-O8	2.50(1)	2.742
Cu-O9W	1.98(1)	2.094
Cu-O10W	2.00(1)	2.079
<Cu-O>	2.15(1)	2.225
C-O		
C-O4	1.27(2)	1.295
C-O6	1.28(2)	1.290
C-O7	1.31(2)	1.280
<C-O>	1.29(2)	1.288
O-H		
O8-H1	1.05	0.989
O9W-H2	0.76	0.987
O9W-H3	0.93	0.989
O10W-H5	1.00	0.990
O10W-H4	0.73	0.975
Hydrogen bonds (Oi-Hj···Ok)		
O8···O3	2.86	2.750
H1···O3	2.04	1.781
O9W···O2	2.76	2.712
H3···O2’	1.86	1.758
O9W···O1	2.77	2.777
H2···O1	2.02	1.792
O10W···O9W	3.23	3.102
H4···O9W	2.70	2.307
O10W···O2	3.05	2.982
H4···O2	2.39	2.292
O10W···O1	2.73	2.743
H5···O1	1.87	1.762

Table S.3. Interatomic angles in roubaultite (in degrees).

Angle	Exp.[1]	Calc.
O-U1-O		
O1-U1-O1’	180.00	180.00
O1-U1-O4	88.10	88.16
O1-U1-O5	90.89	90.05
O1-U1-O6	89.97	89.38
O4-U1-O5	65.29	65.07
O5-U1-O6	65.27	65.15
O6-U1-O4’	49.52	49.85
O-U2-O		
O2-U2-O3	177.20	177.16
O2-U2-O4	88.54	86.41
O2-U2-O5	91.08	90.94
O2-U2-O5’	92.46	92.54
O2-U2-O6	89.96	91.37
O2-U2-O8	86.08	87.29
O4-U2-O5	66.10	65.33
O5-U2-O5’	69.71	69.75
O5’-U2-O6	66.88	67.91
O6-U2-O8	78.25	79.25
O8-U2-O4	79.05	77.77
O-Cu-O		
O8-Cu-O7’	171.59	171.80
O8-Cu-O7	98.76	99.71
O8-Cu-O9W	84.68	77.15
O8-Cu-O8’	84.25	88.48
O7-Cu-O9W	89.82	92.60
O9W-Cu-O8’	88.81	88.93
O8’-Cu-10W	91.96	92.14
O-C-O		
O4-C1-O6	114.81	114.72
O6-C1-O7	123.39	124.17
O7-C1-O4	121.75	121.11
H-O-H		
H2-O9W-H3	98.71	107.21
H4-O10W-H5	112.91	105.41
Hydrogen bonds (Oi-Hj···Ok)		
O8-H1···O3	132.26	165.98
O9W-H3···O2’	162.81	160.93
O9W-H2···O1	166.48	175.09
O10W-H4···O9W	131.37	138.04
O10W-H4···O2	148.88	126.93
O10W-H5···O1	143.09	170.68

Table S.4. Most intense reflections in the X-ray diffraction pattern of roubaultite: (a) X-ray pattern derived from the experimental crystal structure;[1] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	**(a) Experimental[1]**			**(b) Calculated**		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[0 -1 1]	15.918	5.563	100.000	15.779	5.612	-0.14
[0 1 0]	11.406	7.751	35.652	11.335	7.800	-0.07
[-2 1 0]	27.605	3.229	22.332	27.333	3.260	-0.27
[-1 1 2]	28.004	3.184	18.850	27.760	3.211	-0.24
[1 1 2]	29.461	3.030	16.048	29.243	3.052	-0.22
[1 1 1]	21.415	4.146	15.966	21.358	4.157	-0.06
[2 0 0]	25.780	3.453	14.532	25.714	3.462	-0.07
[0 -2 1]	25.431	3.500	14.435	25.242	3.525	-0.19
[0 0 1]	11.273	7.843	14.429	11.155	7.925	-0.12
[-1 2 1]	28.047	3.179	13.478	27.730	3.215	-0.32
[0 -2 2]	32.154	2.782	12.078	31.868	2.806	-0.29
[-2 0 1]	27.801	3.206	11.795	27.730	3.215	-0.07
[1 2 1]	29.739	3.002	11.519	29.698	3.006	-0.04
[1 0 2]	26.548	3.355	11.051	26.242	3.393	-0.31
[-1 1 1]	19.983	4.440	10.884	19.790	4.483	-0.19
[1 0 0]	12.808	6.906	10.603	12.776	6.924	-0.03
[2 1 0]	28.922	3.085	10.232	29.004	3.076	0.08
[-1 0 2]	25.665	3.468	8.364	25.524	3.487	-0.14
[-2 -1 1]	30.674	2.912	7.833	30.779	2.903	0.11
[2 -2 1]	35.801	2.506	7.822	35.262	2.543	-0.54

Table S.5. Interatomic distances in fontanite (in Å).

Distance	Exp.[2]	Calc.
U1-O		
U1-O3	1.77(3)	1.820
U1-O12	1.83(3)	1.813
U1-O7	2.21(3)	2.235
U1-O11	2.23(3)	2.240
U1-O10	2.54(3)	2.560
U1-O6	2.53(3)	2.568
U1-O9	2.56(3)	2.531
U1-O13	2.57(3)	2.540
<U1-Oap>	1.80(3)	1.817
<U1-Oeq>	2.44(3)	2.446
U2-O		
U2-O5	1.73(3)	1.817
U2-O20	1.73(3)	1.834
U2-O11	2.19(3)	2.227
U2-O7	2.21(3)	2.222
U2-O2	2.36(2)	2.370
U2-O9	2.44(3)	2.459
U2-O10	2.44(3)	2.429
<U2-Oap>	1.73(3)	1.826
<U2-Oeq>	2.33(3)	2.341
U3-O		
U3-O1	1.73(3)	1.834
U3-O14	1.76(3)	1.818
U3-O7	2.28(3)	2.238
U3-O11	2.28(3)	2.239
U3-O8	2.40(3)	2.339
U3-O6	2.42(3)	2.435
U3-O13	2.45(2)	2.457
<U1-Oap>	1.75(3)	1.826
<U2-Oeq>	2.37(3)	2.342
Ca-O		
Ca-O17W	2.42(4)	2.517
Ca-O15W	2.47(4)	2.635
Ca-O3	2.49(3)	2.417
Ca-O18W	2.45(4)	2.438
Ca-O5	2.52(4)	2.512
Ca-O4W	2.56(4)	2.452
Ca-O19W	2.56(5)	2.663
Ca-O16W	2.57(4)	2.351
<Ca-O>	2.51(4)	2.498
C1-O		
C1-O8	1.28(4)	1.267
C1-O9	1.27(4)	1.300
C1-O10	1.33(4)	1.300
<C1-O>	1.29(4)	1.289
C2-O		
C2-O13	1.27(4)	1.297
C2-O6	1.31(4)	1.296
C2-O2	1.30(3)	1.274
<C2-O>	1.29(4)	1.289
O-H		
O4W-H1	-	0.971
O4W-H2	-	0.978
O15W-H3	-	0.972
O15W-H4	-	0.987
O16w-H5	-	0.976
O16w-H6	-	0.991
O17w-H7	-	0.977
O17w-H8	-	0.980
O18w-H9	-	0.972
O18w-H10	-	0.975
O19w-H11	-	0.983
O19w-H12	-	0.986

Table S.6. Interatomic angles in fontanite (in degrees).

Angle	Exp.[2]	Calc.
O-U1-O		
O3-U1-O12	177.63	178.52
O3-U1-O7	91.12	88.37
O3-U1-O11	86.90	90.84
O3-U1-O10	93.29	88.99
O3-U1-O6	88.56	93.47
O3-U1-O9	91.30	89.74
O3-U1-O13	90.80	90.92
O-U2-O		
O5-U2-O20	172.88	174.01
O5-U2-O11	92.77	93.50
O5-U2-O7	91.01	90.85
O5-U2-O2	86.14	86.13
O5-U2-O9	88.62	88.17
O5-U2-O10	90.07	89.93
O-U3-O		
O1-U3-O14	177.26	175.28
O1-U3-O7	90.55	90.52
O1-U3-O11	89.39	91.01
O1-U3-O8	89.33	85.32
O1-U3-O6	93.70	90.11
O1-U3-O13	89.91	89.99
O-Ca-O		
O17W-Ca-O15W	75.31	69.95
O17W-Ca-O3	79.27	78.25
O17W-Ca-O18W	145.75	149.31
O17W-Ca-O5	104.28	121.42
O17W-Ca-O4W	74.84	76.37
O17W-Ca-O19W	141.76	125.54
O17W-Ca-O16W	75.94	73.46
O-C1-O		
O8-C1-O9	123.94	123.27
O9-C1-O10	115.99	113.59
O10-C1-O8	120.07	123.14
O-C2-O		
O13-C2-O6	116.54	114.15
O6-C2-O2	119.27	122.86
O2-C2-O13	124.00	122.99
H-O-H		
H1-O4W-H2	-	104.89
H3-O15W-H4	-	105.27
H5-O16W-H6	-	104.69
H7-O17W-H8	-	104.38
H9-O18W-H10	-	107.21
H11-O19W-H12	-	106.38

Table S.7. Hydrogen bond parameters (O1-H···O2) in fontanite (distances in Å and angles in degrees).

Hydrogen bond	O1···O2	H···O2	α(O1-H···O2)
O4W-H1···O1	2.977	2.207	135.42
O4W-H2···O4W	2.785	2.734	104.53
O15W-H3···O1	2.957	2.134	141.47
O15W-H4···O20	2.723	1.743	171.43
O16W-H5···O14	2.825	1.874	164.25
O16W-H6···O19W	2.705	1.716	174.82
O17W-H7···O20	2.896	1.960	159.41
O17W-H8···O1	2.824	1.875	162.21
O18W-H9···O14	3.097	2.161	161.16
O18W-H10···O2	2.938	2.016	156.81
O19W-H12···O12	2.739	1.757	173.19

Table S.8. Most intense reflections in the X-ray diffraction pattern of fontanite: (a) X-ray pattern derived from the experimental crystal structure;[2] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	(a) Experimental[2]			(b) Calculated		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[0 2 0]	10.232	8.638	100.000	10.163	8.697	-0.07
[0 1 2]	12.592	7.024	22.670	12.665	6.984	0.07
[2 2 0]	27.585	3.231	18.136	27.561	3.234	-0.02
[2 0 0]	25.547	3.484	17.287	25.550	3.484	0.00
[0 3 1]	16.424	5.393	16.476	16.342	5.420	-0.08
[1 1 5]	32.222	2.776	14.026	32.293	2.770	0.07
[-1 2 3]	23.857	3.727	11.816	24.049	3.698	0.19
[-1 1 5]	32.196	2.778	10.929	32.559	2.748	0.36
[0 4 0]	20.547	4.319	10.800	20.406	4.349	-0.14
[1 2 3]	23.878	3.724	10.402	23.838	3.730	-0.04
[0 1 1]	7.691	11.486	9.147	7.703	11.468	0.01
[0 0 2]	11.500	7.689	8.769	11.595	7.626	0.10
[-1 0 3]	21.493	4.131	8.313	21.738	4.085	0.25
[0 3 2]	19.241	4.609	7.716	19.215	4.616	-0.03

Table S.9. Interatomic distances in sharpite (in Å).

Distance	Exp.[3]	Calc.
U1-O		
U1-O2	1.756(15)	1.790
U1-O5	1.764(17)	1.797
U1-O1	2.469(18)	2.499
U1-O1’	2.469(18)	2.499
U1-O4	2.531(9)	2.482
U1-O4’	2.531(9)	2.482
U1-O6	2.405(12)	2.713
U1-O6’	2.405(12)	2.713
<U1-Oap>	1.760(17)	1.794
<U1-Oeq>	2.468(18)	2.565
U2-O		
U2-O3	1.769(19)	1.825
U2-O3’	1.769(19)	1.825
U2-O6	2.442(11)	2.462
U2-O6’	2.442(11)	2.462
U2-O6’’	2.442(11)	2.462
U2-O6’’’	2.442(11)	2.462
U2-O9	2.4516(4)	2.492
U2-O9’	2.4516(4)	2.492
<U2-Oap>	1.769(19)	1.825
<U2-Oeq>	2.445(11)	2.472
Ca-O		
Ca1-O4	2.415(10)	2.442
Ca1-O4’	2.415(10)	2.442
Ca1-O4’’	2.415(10)	2.442
Ca1-O4’’’	2.415(10)	2.442
Ca1-O7W	2.744(14)	2.835
Ca1-O7W’	2.744(14)	2.835
Ca1-O8W	2.380(2)	2.453
Ca1-O8W’	2.380(2)	2.453
<Ca1-O>	2.489(10)	2.543
C1-O		
C1-O1	1.340(2)	1.311
C1-O4	1.258(13)	1.280
C1-O4’	1.258(13)	1.280
<C1-O>	1.285(13)	1.290
C2-O		
C2-O6	1.293(16)	1.221
C2-O6’	1.293(4)	1.221
C2-O9	1.640(4)	1.613
<C2-O>	1.409(16)	1.352
O-H		
O7W-H7	-	0.985
O7W-H7’	-	0.985
O8W-H8	-	0.978
O8W-H8’	-	0.978
Hydrogen bonds (Oi-Hj···Ok)		
O7W···O5	-	2.826
H7···O5	-	1.878
O8W···O3	-	3.038
H8···O3	-	2.087

Table S.10. Interatomic angles in sharpite (in degrees).

Angle	Exp.[3]	Calc.
O-U1-O		
O2-U1-O5	178.82	177.83
O2-U1-O1	90.42	90.13
O2-U1-O1’	90.42	90.13
O2-U1-O4	88.91	88.38
O2-U1-O4’	88.91	88.38
O2-U1-O6	91.25	92.38
O2-U1-O6’	91.25	92.38
O-U2-O		
O3-U2-O3’	180.00	180.00
O3-U2-O6	91.65	94.63
O3-U2-O6’	91.65	94.63
O3-U2-O6’’	88.35	85.37
O3-U2-O6’’’	88.35	85.37
O3-U2-O9	90.00	90.00
O3-U2-O9’	90.00	90.00
O-Ca-O		
O4-Ca1-O4’	113.24	108.95
O4-Ca1-O4’’	65.93	67.09
O4-Ca1-O4’’’	78.61	73.39
O4-Ca1-O7W	137.20	139.72
O4-Ca1-O7W’	76.24	77.97
O4-Ca1-O8W	79.32	74.41
O4-Ca1-O8W’	146.74	142.77
O-C1-O		
O1-C1-O4	115.14	116.82
O4-C1-O4’	129.65	126.35
O4’-C1-O1	115.14	116.82
O-C2-O		
O6-C2-O6’	138.29	137.35
O6’-C2-O9	110.84	111.29
O9-C2-O6	110.84	111.29
H-O-H		
H7-O7W-H7’	-	99.61
H8-O8W-H8’	-	105.11
Hydrogen bonds (Oi-Hj···Ok)		
O7W-H7···O5	-	160.54
O8W-H8···O3	-	163.46

Table S.11. Most intense reflections in the X-ray diffraction pattern of sharpite: (a) X-ray pattern derived from the experimental crystal structure;[3] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	(a) Experimental[3]			(b) Calculated		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[0 2 4]	19.689	4.505	100.000	19.598	4.526	-0.09
[0 0 2]	8.016	11.021	55.258	7.981	11.068	-0.04
[1 3 4]	29.780	2.998	48.187	29.506	3.025	-0.27
[1 1 1]	19.378	4.577	36.008	19.091	4.645	-0.29
[1 1 3]	22.511	3.947	31.381	22.237	3.995	-0.27
[2 2 4]	41.919	2.153	27.047	41.328	2.183	-0.59
[0 2 3]	16.538	5.356	25.335	16.460	5.381	-0.08
[1 3 3]	27.744	3.213	23.555	27.470	3.244	-0.27
[1 5 0]	33.954	2.638	21.459	33.663	2.660	-0.29
[1 1 2]	20.606	4.307	20.407	20.325	4.366	-0.28
[1 1 0]	18.952	4.679	19.697	18.662	4.751	-0.29
[0 2 2]	13.869	6.380	19.296	13.802	6.411	-0.07
[1 7 4]	47.616	1.908	18.788	47.277	1.921	-0.34
[1 5 8]	47.689	1.906	18.141	47.365	1.918	-0.32
[1 1 7]	34.300	2.612	17.355	34.029	2.633	-0.27
[2 0 0]	36.625	2.452	15.284	36.014	2.492	-0.61
[0 4 0]	22.711	3.912	15.000	22.593	3.932	-0.12
[0 2 0]	11.300	7.825	11.948	11.241	7.865	-0.06
[0 4 7]	36.604	2.453	11.787	36.429	2.464	-0.17
[0 2 6]	26.788	3.325	10.708	26.667	3.340	-0.12
[2 0 8]	49.743	1.832	9.921	49.165	1.852	-0.58
[0 6 7]	45.102	2.009	9.792	44.873	2.018	-0.23
[0 4 3]	25.779	3.453	9.645	25.649	3.470	-0.13
[1 3 2]	26.201	3.399	9.404	25.925	3.434	-0.28
[2 4 0]	43.530	2.077	9.001	42.935	2.105	-0.59
[0 4 8]	39.992	2.253	8.853	39.801	2.263	-0.19
[2 2 6]	45.954	1.973	8.429	45.372	1.997	-0.58
[0 4 2]	24.120	3.687	7.862	23.996	3.706	-0.12
[0 4 1]	23.071	3.852	7.486	22.951	3.872	-0.12
[0 0 8]	32.471	2.755	7.249	32.327	2.767	-0.14
[2 4 1]	43.732	2.068	7.175	43.138	2.095	-0.59

Table S.12. Interatomic distances in widenmannite (in Å).

Distance	Exp.[4]	Calc.
U1-O		
U1-O3 × 2	1.802(19)	1.784
U1-O1 × 2	2.4895(11)	2.485
U1-O2 × 2	2.43(3)	2.466
U1-O4 × 2	2.46(3)	2.464
<U1-Oap>	1.80(2)	1.784
<U1-Oeq>	2.46(3)	2.472
Pb-O		
Pb-O6	2.35(4)	2.303
Pb-O6' × 2	2.683(16)	2.607
Pb-O2 × 2	3.089(17)	3.117
Pb-O3 × 2	3.42(4)	3.457
Pb-O4 × 2	2.781(11)	2.789
Pb-O5	2.574(18)	2.625
Pb-O1	3.37(2)	3.499
<Pb-O>	2.93(4)	2.942
C1-O		
C1-O1	1.30(4)	1.321
C1-O2 × 2	1.29(3)	1.278
<C1-O>	1.29(4)	1.292
C2-O		
C2-O5	1.30(4)	1.273
C2-O4 × 2	1.31(3)	1.305
<C2-O>	1.31(4)	1.294
O-H		
O6-H1	0.819	0.970
Hydrogen bonds (Oi-Hj···Ok)		
O6···O2	3.190	3.294
H1···O2	2.564	2.679
O6···O4	3.200	3.422
H1···O4	3.162	3.116

Table S.13. Interatomic angles in widenmannite (in degrees).

Angle	Exp.[4]	Calc.
O-U1-O		
O3-U1-O3’	179.26	177.368
O3-U1-O1 × 2	90.01	90.087
O3-U1-O2 × 2	90.31	91.104
O3-U1-O4 × 2	89.67	88.822
O1-U1-O4	64.089	67.324
O4-U1-O4’	54.999	52.597
O4’-U1-O1’	64.089	67.324
O1’-U1-O2	54.203	53.523
O2-U1-O2’	68.419	65.949
O2’-U1-O1	54.203	53.223
O-Pb-O		
O6-Pb-O6’	76.07	75.973
O6-Pb-O2 × 2	135.28	130.632
O6-Pb-O3 × 2	125.14	128.728
O6-Pb-O4 × 2	76.68	86.854
O6-Pb-O5	79.02	74.683
O6-Pb-O1	116.56	125.453
O-C1-O		
O1-C1-O2	119.94	117.121
O2-C1-O2’	120.11	125.759
O2’-C1-O1	119.94	117.121
O-C2-O		
O5-C1-O4	119.96	122.670
O4-C1-O4’	120.08	114.660
O4’-C1-O5	119.96	122.670
Hydrogen bonds (Oi-Hj···Ok)		
O6-H1···O2	134.198	121.768
O6-H1’···O4	85.270	100.084

Table S.14. Most intense reflections in the X-ray diffraction pattern of widenmannite: (a) X-ray pattern derived from the experimental crystal structure;[4] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	(a) Experimental[4]			(b) Calculated		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[1 2 1]	27.940	3.191	100.000	27.716	3.216	-0.22
[0 2 1]	21.355	4.158	82.522	21.012	4.225	-0.34
[1 0 2]	26.752	3.330	74.786	26.621	3.346	-0.13
[1 1 0]	20.178	4.397	36.844	20.158	4.402	-0.02
[1 2 4]	48.571	1.873	31.106	48.037	1.893	-0.53
[2 0 0]	36.063	2.489	29.738	36.193	2.480	0.13
[0 2 2]	27.502	3.241	29.629	27.104	3.287	-0.40
[0 4 0]	38.324	2.347	28.495	37.648	2.387	-0.68
[1 4 2]	47.354	1.918	25.718	46.703	1.943	-0.65
[1 1 1]	22.506	3.947	24.154	22.436	3.960	-0.07
[1 0 1]	20.396	4.351	22.702	20.394	4.351	0.00
[2 2 1]	42.293	2.135	22.464	42.223	2.139	-0.07
[0 0 3]	29.894	2.987	19.784	29.538	3.022	-0.36
[2 2 2]	45.944	1.974	17.474	45.796	1.980	-0.15
[1 0 3]	35.011	2.561	16.591	34.736	2.581	-0.28
[0 2 3]	35.602	2.520	15.040	35.117	2.553	-0.48
[0 1 2]	21.967	4.043	13.472	21.686	4.095	-0.28
[2 1 2]	42.628	2.119	13.321	42.587	2.121	-0.04
[0 1 1]	13.652	6.481	12.355	13.457	6.575	-0.19
[2 4 0]	53.639	1.707	11.430	53.215	1.720	-0.42
[0 1 3]	31.407	2.846	8.798	31.017	2.881	-0.39
[2 0 3]	47.521	1.912	8.134	47.385	1.917	-0.14
[0 1 4]	41.409	2.179	8.108	40.894	2.205	-0.52
[1 1 2]	28.419	3.138	7.823	28.239	3.158	-0.18
[2 2 3]	51.578	1.771	7.318	51.317	1.779	-0.26
[2 1 3]	48.559	1.873	7.087	48.391	1.880	-0.17

Table S.15. Interatomic distances in grimselite (in Å).

Distance	Exp.[5]	Calc.
	U-O	
U-O3	1.785(8)	1.822
U-O3’	1.785(8)	1.824
U-O2	2.425(4)	2.435
U-O2’	2.425(4)	2.442
U-O2’’	2.43(2)	2.488
U-O4	2.408(7)	2.405
U-O4’	2.408(8)	2.409
U-O4’’	2.41(1)	2.422
<U-Oap>	1.785(8)	1.823
<U-Oeq>	2.418(8)	2.434
	K-O	
K-O1	2.809(9)	2.722
K-O1’	2.809(9)	2.731
K-O2	2.730(6)	2.689
K-O2’	2.730(7)	2.698
K-O4	2.869(5)	2.778
K-O4’	2.869(7)	2.796
K-O5W	2.925(8)	3.060
K-O5W’	2.925(8)	3.064
<K-O>	2.826(9)	2.817
	Na-O	
Na-O1	2.945(4)	2.872
Na-O1’	2.945(4)	2.919
Na-O1’’	2.95(2)	2.956
Na-O3	2.362(8)	2.385
Na-O3’	2.362(8)	2.391
Na-O4	2.45(1)	2.463
Na-O4’	2.45(1)	2.522
Na-O4’’	2.45(1)	2.538
<Na-O>	2.615(8)	2.631
	C-O	
C-O1	1.35(2)	1.318
C-O2	1.30(2)	1.301
C-O4	1.25(1)	1.271
<C-O>	1.298(2)	1.297
	O-H	
O5W-H1	-	0.980
O5W-H1’	-	0.976

Table S.16. Hydrogen bond parameters (O1-H···O2) in grimselite (distances in Å and angles in degrees).

Hydrogen bond	O1···O2	H···O2	α(O1-H···O2)
O5W-H1···O4	3.039	2.092	161.748
O5W-H1'···O4'	3.162	2.242	156.285

Table S.17. Most intense reflections in the X-ray diffraction pattern of grimselite: (a) X-ray pattern derived from the experimental crystal structure;[5] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	**(a) Experimental[5]**			**(b) Calculated**		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[1 0 1]	15.36	5.77	100.00	15.08	5.87	-0.28
[1 0 0]	10.98	8.06	42.03	11.11	7.96	0.13
[1 1 2]	28.91	3.09	41.48	28.52	3.13	-0.39
[3 0 0]	33.37	2.69	31.23	33.76	2.65	0.39
[1 1 0]	19.08	4.65	28.15	18.93	4.68	-0.15
[2 1 1]	31.31	2.86	27.94	31.18	2.87	-0.13
[2 0 1]	24.59	3.62	27.29	24.76	3.59	0.17
[2 2 2]	44.72	2.03	21.05	44.26	2.05	-0.46
[1 0 2]	24.22	3.68	20.33	23.93	3.72	-0.29
[0 0 2]	21.52	4.13	18.12	21.09	4.21	-0.43
[2 2 0]	38.72	2.33	15.05	38.40	2.34	-0.32
[3 1 1]	41.89	2.16	14.53	41.90	2.15	0.01
[1 0 3]	34.42	2.61	10.46	33.89	2.64	-0.53
[2 0 0]	22.07	4.03	10.38	22.32	3.98	0.25
[3 2 1]	50.61	1.80	10.22	50.39	1.81	-0.22
[2 1 0]	29.33	3.04	9.39	28.92	3.08	-0.41
[3 1 2]	46.20	1.97	8.45	46.74	1.94	0.54
[3 0 4]	56.19	1.64	8.34	55.81	1.65	-0.38

Table S.18. Interatomic distances in čejkaite (in Å).

Distance	Exp.[6]	Calc.
U-O		
U-O10	1.841(11)	1.840
U-O11	1.885(11)	1.840
U-O2	2.377(17)	2.374
U-O1	2.402(17)	2.381
U-O9’	2.434(15)	2.389
U-O5	2.447(15)	2.389
U-O7	2.426(13)	2.389
U-O6	2.421(15)	2.390
<U-Oap>	1.86	1.840
<U-Oeq>	2.42	2.385
Na1-O		
Na1-O4	2.39(3)	2.418
Na1-O3	2.40(3)	2.434
Na1-O8	2.41(3)	2.396
Na1-O3’	2.52(2	2.517
Na1-O4’	2.55(3)	2.538
Na1-O8’	2.62(3)	2.606
<Na1-O>	2.48	2.485
Na2-O		
Na2-O6	2.313(17)	2.378
Na2-O8	2.656(15)	2.345
Na2-O2	2.31(2)	2.331
Na2-O11	2.33(2)	2.540
Na2-O10	2.443(18)	2.434
Na2-O5	2.656(15)	2.658
<Na2-O>	2.39	2.448
Na3-O		
Na3-O5	2.251(17)	2.341
Na3-O3	2.314(18)	2.340
Na3-O9	2.333(18)	2.372
Na3-O11	2.501(19)	2.487
Na3-O10	2.506(19)	2.472
Na3-O2	2.918(17)	2.897
<Na3-O>	2.47	2.485
Na4-O		
Na4-O4	2.292(19)	2.339
Na4-O7	2.335(16)	2.353
Na4-O1	2.340(19)	2.347
Na4-O10	2.507(19)	2.475
Na4-O11	2.561(19)	2.501
Na4-O1’	2.794(16)	2.792
<Na4-O>	2.47	2.468
C1-O		
C1-O3	1.22(3)	1.261
C1-O6	1.29(3)	1.305
C1-O1	1.34(3)	1.318
<C1-O>	1.28	1.295
C2-O		
C2-O4	1.23(2)	1.260
C2-O2	1.27(3)	1.315
C2-O9	1.35(3)	1.306
<C2-O>	1.28	1.294
C3-O		
C3-O8	1.21(3)	1.259
C3-O7	1.30(3)	1.307
C3-O5	1.37(3)	1.317
<C3-O>	1.29	1.294

Table S.19. Most intense reflections in the X-ray diffraction pattern of čejkaite: (a) X-ray pattern derived from the experimental crystal structure;[6] (b) X-ray pattern derived from the calculated crystal structure.

$[hkl]$	(a) Experimental[6]			(b) Calculated		
	2θ (°)	d (Å)	I (%)	2θ (°)	d (Å)	$\Delta(2\theta)$
[1 1 1]	17.806	4.977	100.000	17.675	5.014	-0.13
[1 3 0]	19.084	4.647	94.622	19.043	4.657	-0.04
[0 2 1]	17.620	5.030	85.711	17.516	5.059	-0.10
[-1 1 1]	17.438	5.082	72.782	17.349	5.108	-0.09
[0 0 2]	27.674	3.221	70.550	27.437	3.248	-0.24
[1 1 0]	10.988	8.046	66.692	10.957	8.068	-0.03
[2 0 0]	19.093	4.645	47.345	19.033	4.659	-0.06
[3 3 0]	33.383	2.682	38.530	33.287	2.690	-0.10
[0 2 0]	10.983	8.050	33.663	10.962	8.065	-0.02
[0 4 1]	26.085	3.413	28.981	25.987	3.426	-0.10
[2 2 1]	26.346	3.380	28.818	26.202	3.398	-0.14
[-2 -2 1]	25.840	3.445	28.214	25.753	3.457	-0.09
[2 6 0]	38.725	2.323	27.234	38.638	2.328	-0.09
[-2 -4 1]	32.320	2.768	24.668	32.227	2.775	-0.09
[-3 -1 1]	32.225	2.776	24.378	32.126	2.784	-0.10
[1 5 1]	32.626	2.742	22.067	32.509	2.752	-0.12
[-1 5 1]	32.419	2.760	21.586	32.325	2.767	-0.09
[3 1 1]	32.846	2.725	19.734	32.676	2.738	-0.17
[2 2 0]	22.078	4.023	19.709	22.016	4.034	-0.06
[0 6 0]	33.367	2.683	19.371	33.303	2.688	-0.06
[2 4 1]	32.735	2.734	18.093	32.594	2.745	-0.14
[1 9 0]	52.021	1.757	15.390	51.915	1.760	-0.11
[4 6 0]	52.040	1.756	15.344	51.896	1.761	-0.14
[-4 -2 1]	42.530	2.124	15.261	42.406	2.130	-0.12
[1 1 3]	43.833	2.064	15.078	43.439	2.082	-0.39
[-3 -5 1]	42.602	2.121	14.997	42.489	2.126	-0.11
[5 3 0]	52.051	1.756	14.993	51.884	1.761	-0.17
[4 0 0]	38.744	2.322	13.636	38.619	2.330	-0.13
[1 1 2]	30.080	2.969	13.540	29.822	2.994	-0.26
[-1 7 1]	42.755	2.113	13.443	42.647	2.118	-0.11
[1 7 1]	42.919	2.106	12.185	42.792	2.112	-0.13
[3 5 1]	43.094	2.097	11.639	42.925	2.105	-0.17
[4 2 1]	43.185	2.093	10.563	42.986	2.102	-0.20
[0 2 3]	43.590	2.075	10.300	43.225	2.091	-0.37
[0 2 2]	29.855	2.990	10.237	29.626	3.013	-0.23
[0 4 0]	22.068	4.025	9.856	22.026	4.032	-0.04
[2 2 3]	48.437	1.878	9.664	48.022	1.893	-0.41
[0 4 3]	47.983	1.895	9.164	47.629	1.908	-0.35
[0 0 4]	57.152	1.610	9.081	56.628	1.624	-0.52
[-2 4 3]	51.672	1.768	8.700	51.361	1.778	-0.31
[3 3 2]	44.374	2.040	8.545	44.086	2.053	-0.29
[-3 1 3]	51.465	1.774	7.889	51.166	1.784	-0.30
[1 5 3]	52.304	1.748	7.721	51.930	1.759	-0.37
[2 2 2]	36.061	2.489	7.693	35.790	2.507	-0.27
[5 1 1]	51.820	1.763	7.678	51.587	1.770	-0.23
[-2 2 3]	47.537	1.911	7.557	47.223	1.923	-0.31
[-1 3 2]	33.635	2.662	7.523	33.435	2.678	-0.20
[2 8 1]	51.581	1.771	7.486	51.425	1.776	-0.16
[0 4 2]	35.674	2.515	7.354	35.459	2.530	-0.21
[0 6 2]	43.882	2.062	7.350	43.673	2.071	-0.21
[-3 -7 1]	51.231	1.782	7.196	51.103	1.786	-0.13
[-5 -1 1]	51.108	1.786	7.168	50.956	1.791	-0.15
[3 7 1]	51.658	1.768	7.141	51.481	1.774	-0.18
[-2 2 2]	35.297	2.541	7.048	35.112	2.554	-0.18
[-2 -8 1]	51.296	1.780	7.042	51.173	1.784	-0.12

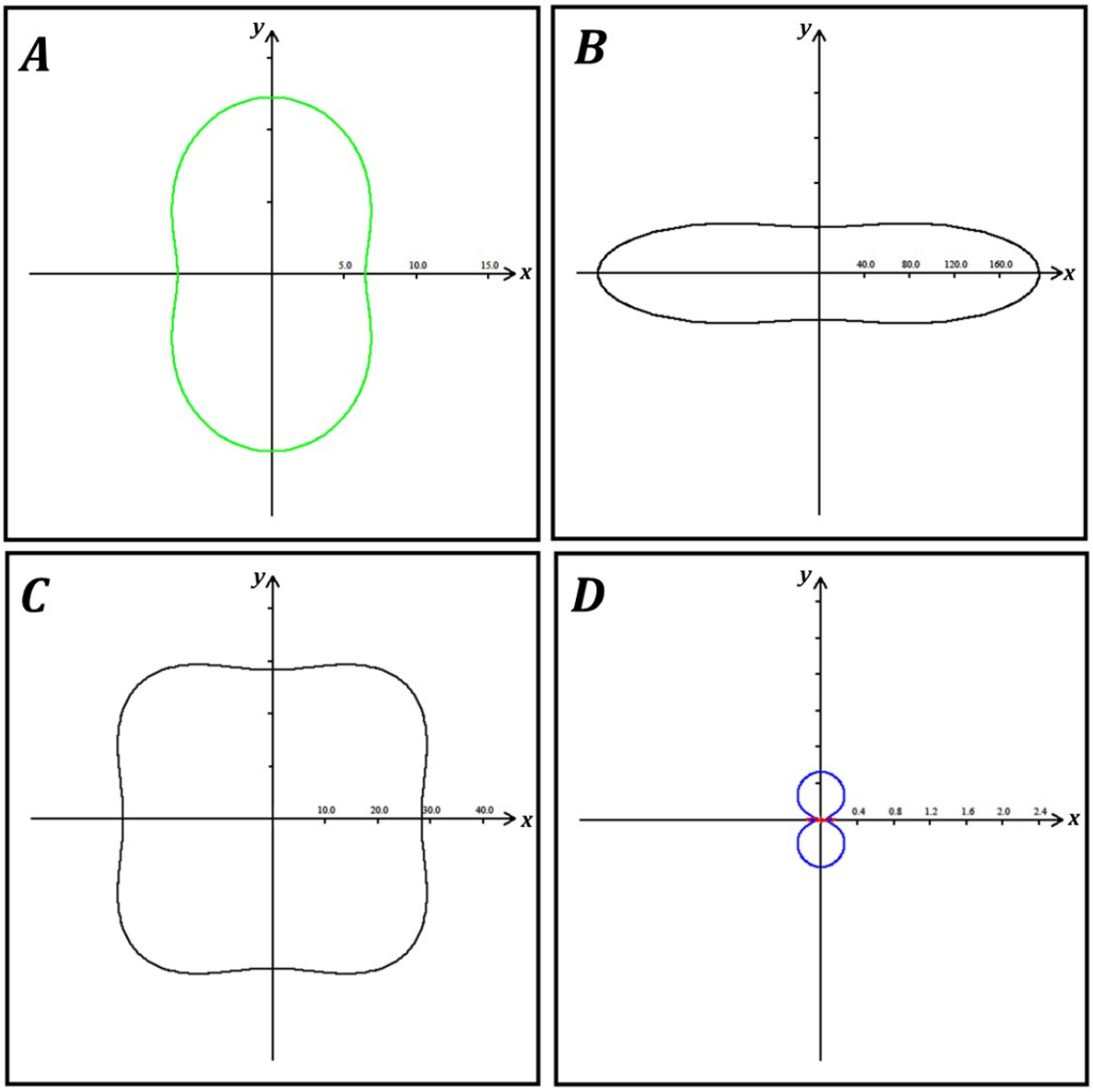

Fig. S.1. Two-dimensional projections in the xy plane of the tridimensional representations of the elastic properties of sharpite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum (blue) and minimum (red) Poisson's ratio. The labels of the marks in the vertical axis are exactly the same as those of the horizontal one.

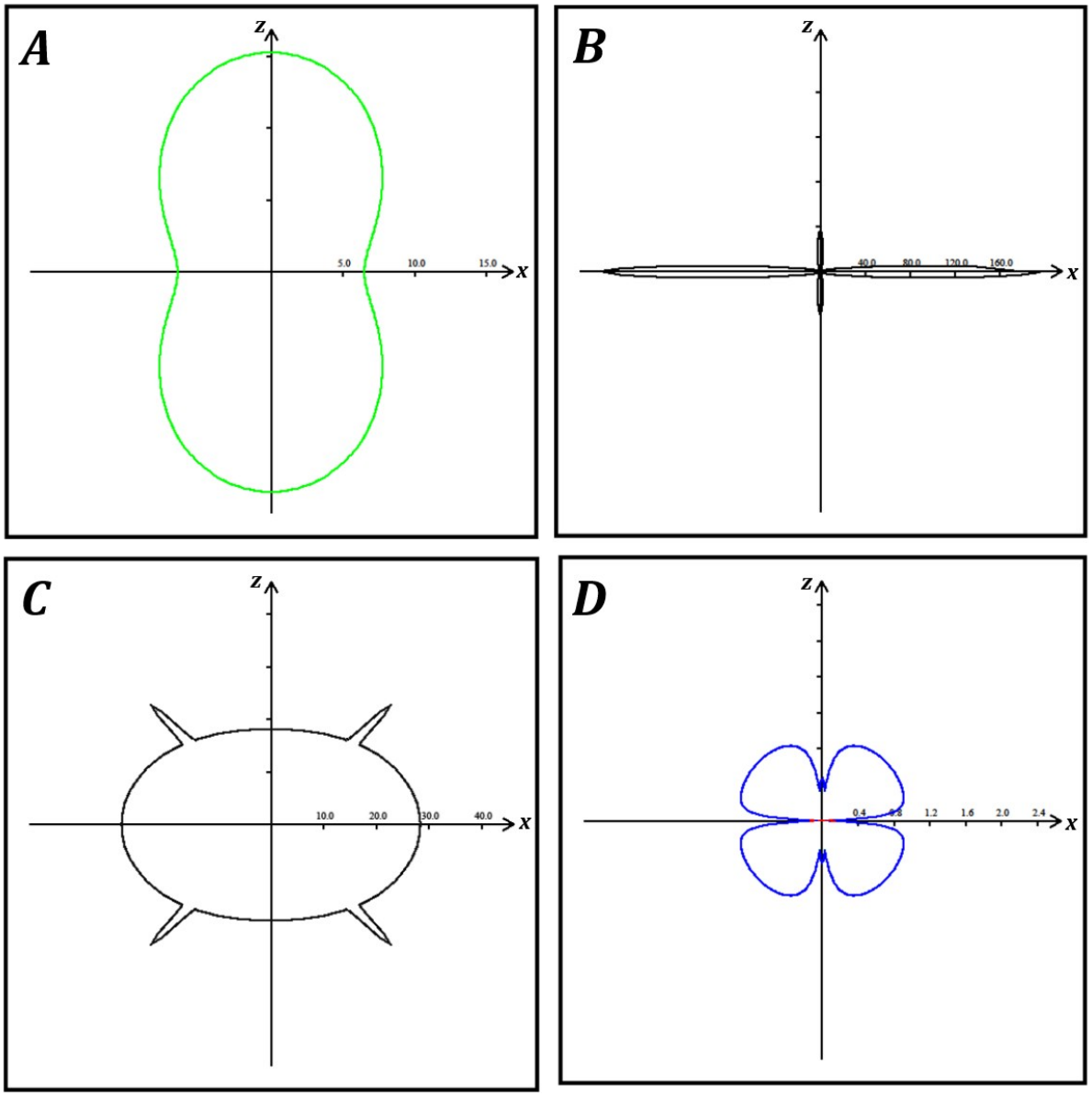

Fig. S.2. Two-dimensional projections in the xz plane of the tridimensional representations of the elastic properties of sharpite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum (blue) and minimum (red) Poisson's ratio.

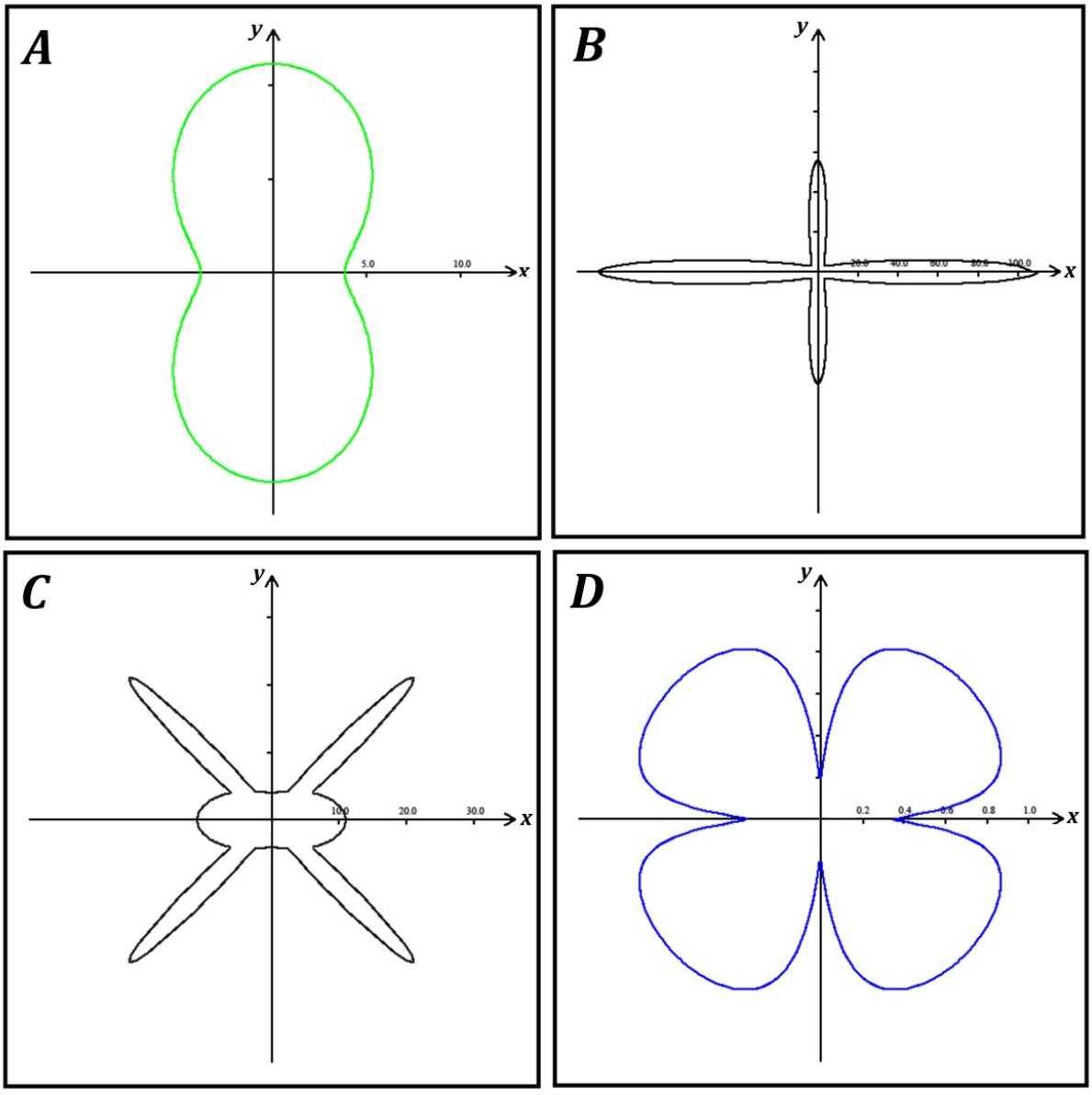

Fig. S.3. Two-dimensional projections in the xy plane of the tridimensional representations of the elastic properties of widenmannite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

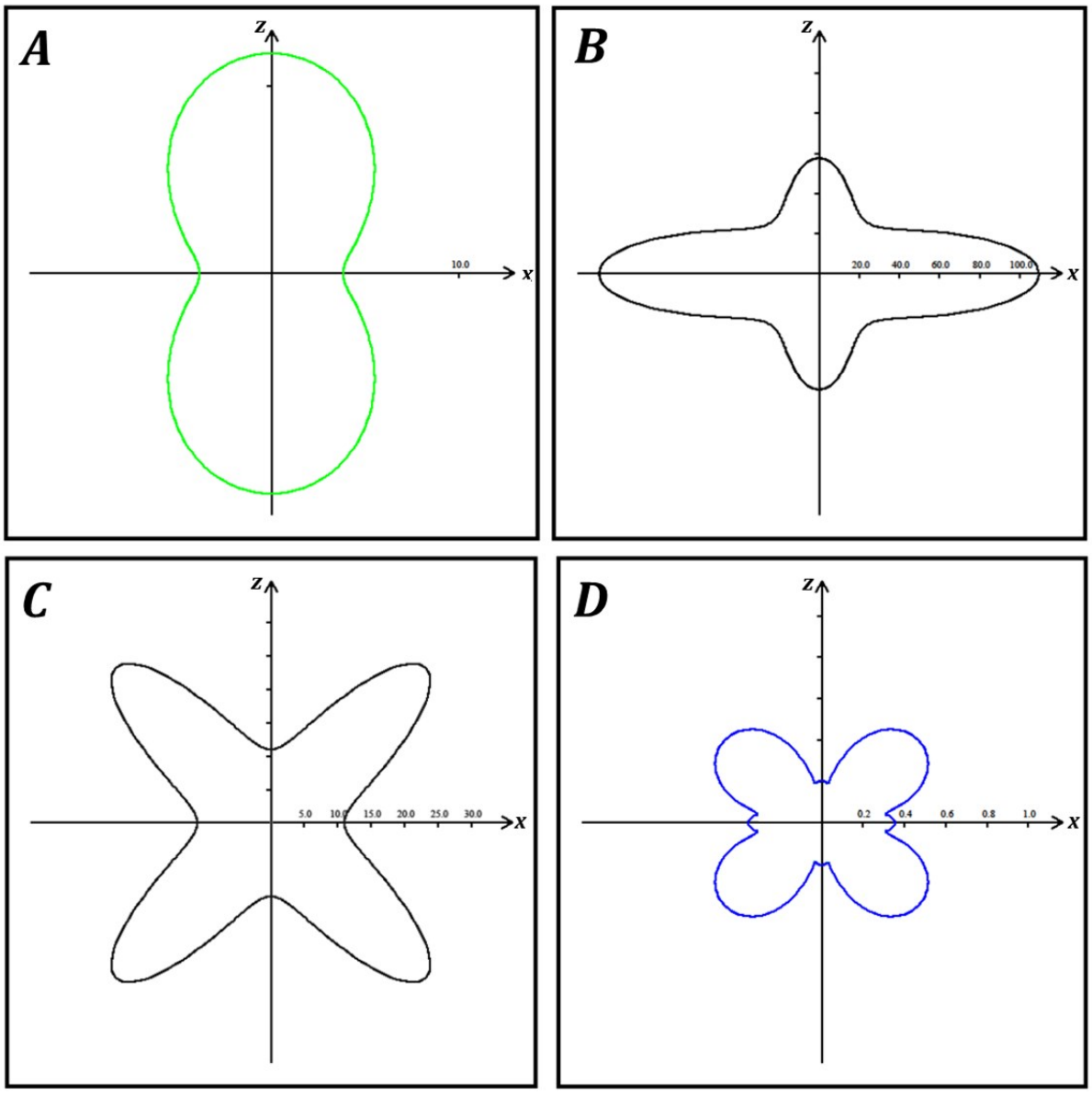

Fig. S.4. Two-dimensional projections in the xz plane of the tridimensional representations of the elastic properties of widenmannite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

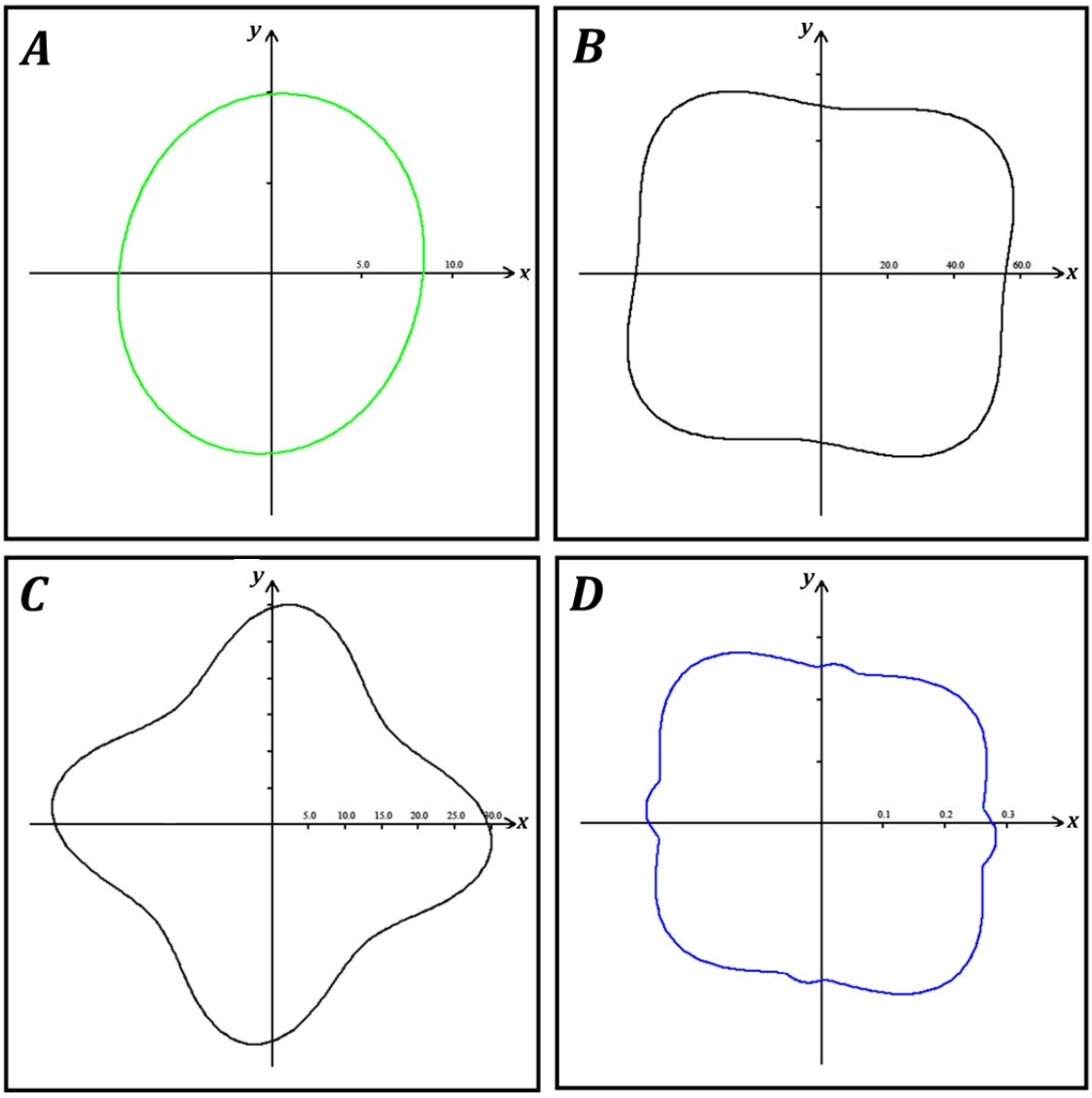

Fig. S.5. Two-dimensional projections in the xy plane of the tridimensional representations of the elastic properties of grimselite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

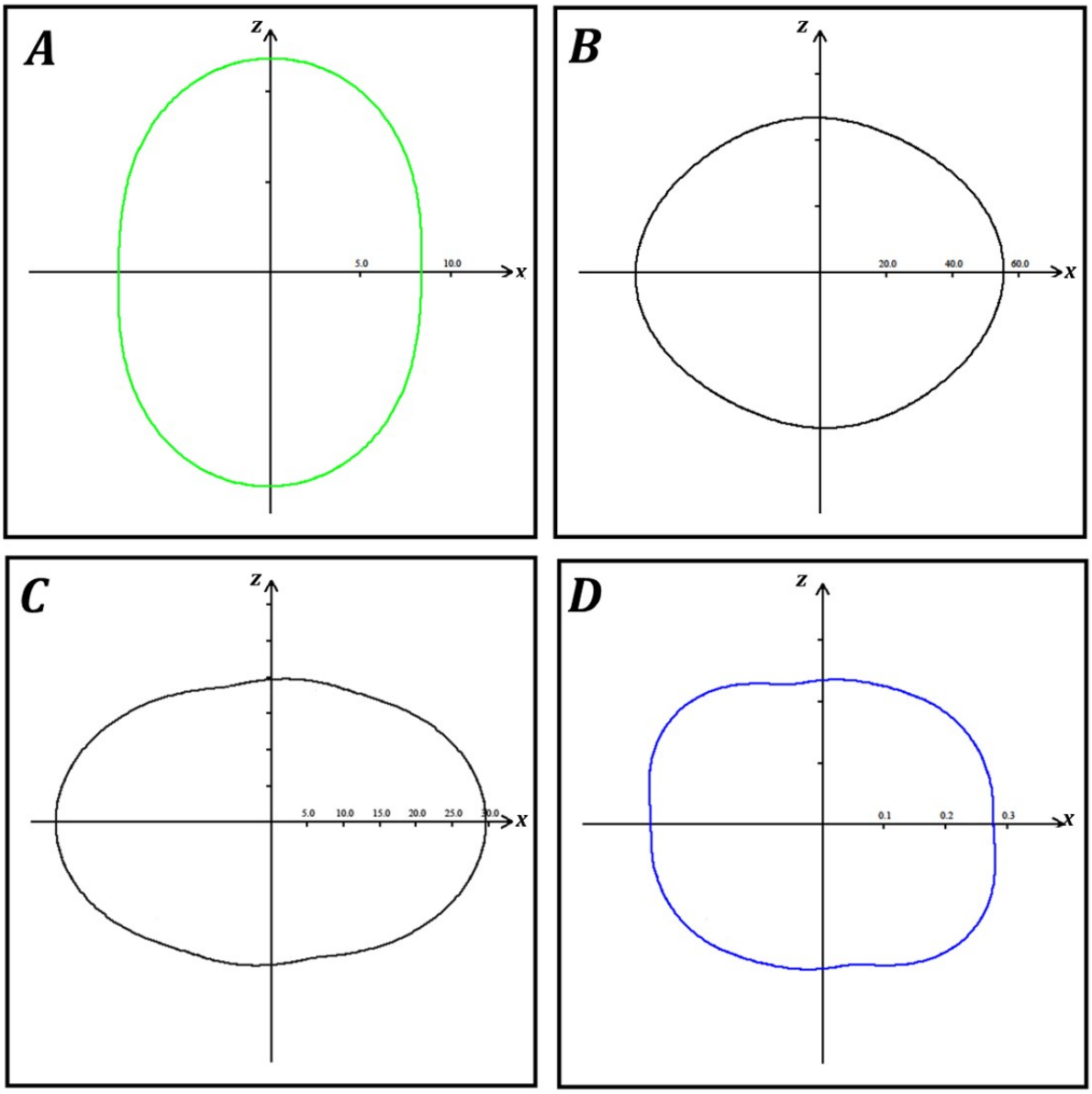

Fig. S.6. Two-dimensional projections in the xz plane of the tridimensional representations of the elastic properties of grimselite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

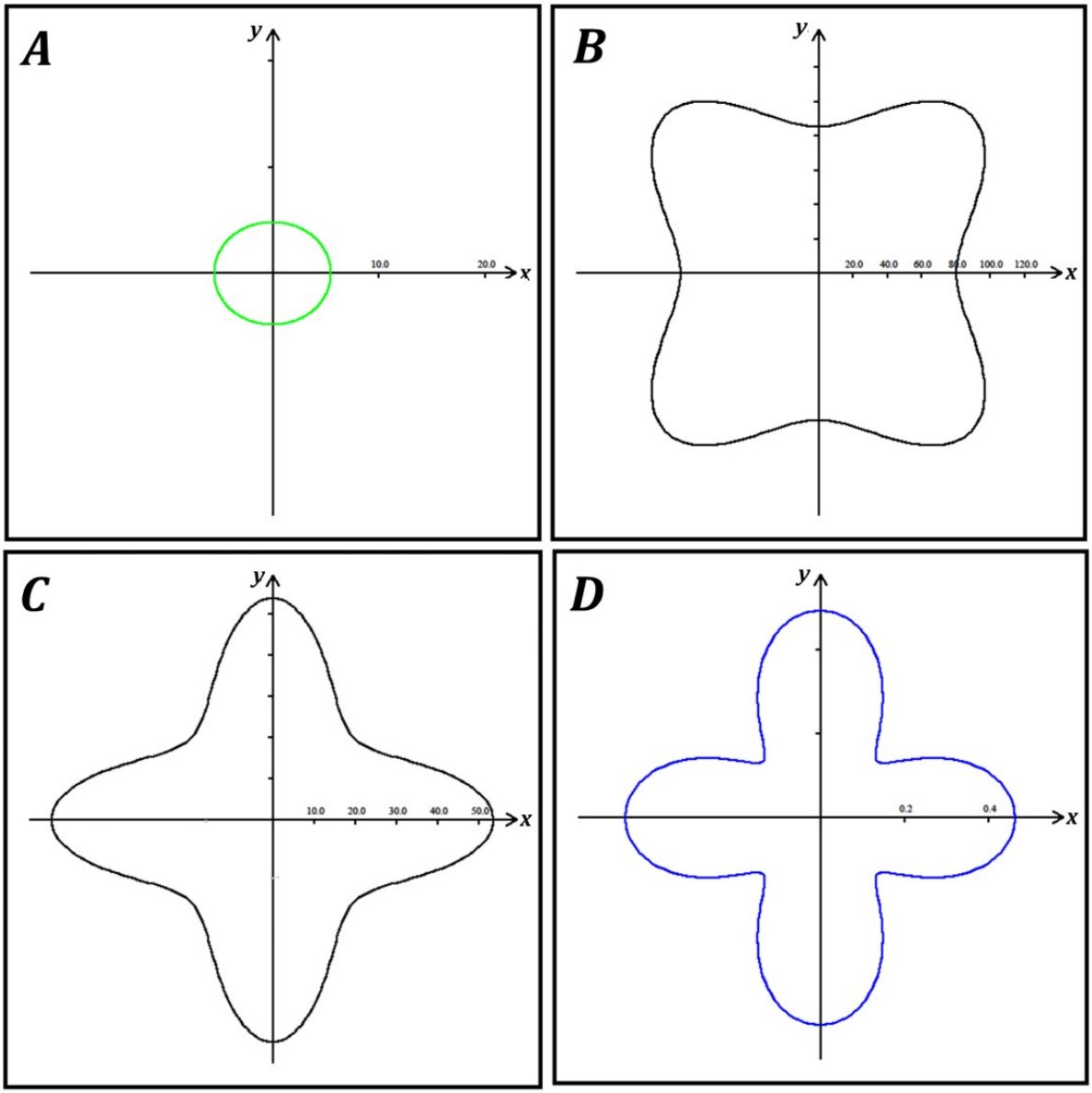

Fig. S.7. Two-dimensional projections in the xy plane of the tridimensional representations of the elastic properties of čejkaite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

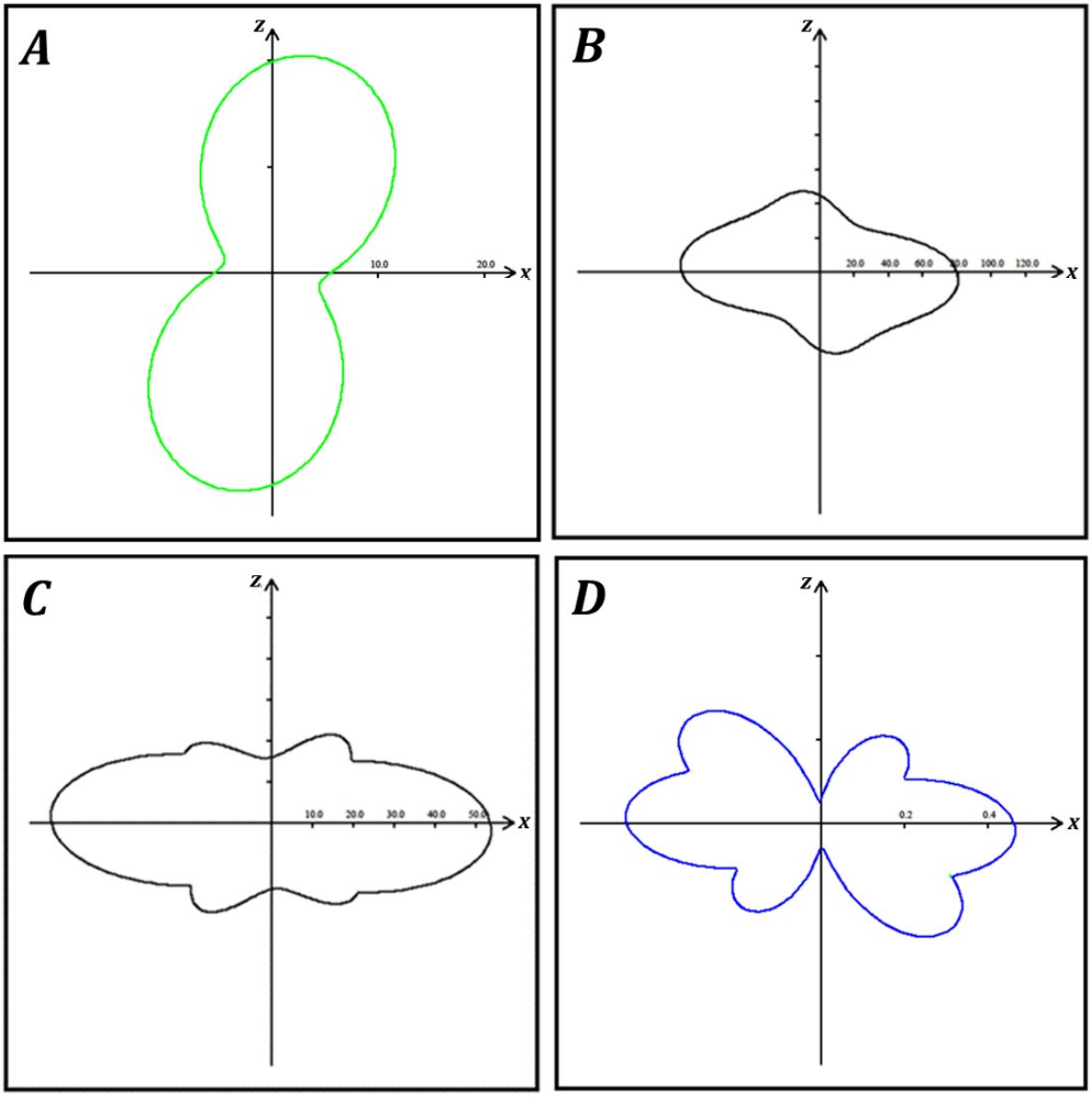

Fig. S.8. Two-dimensional projections in the xz plane of the tridimensional representations of the elastic properties of čejkaite as a function of the orientation of the applied strain: (A) Compressibility; (B) Young modulus; (C) Maximum shear modulus; (D) Maximum Poisson's ratio.

Table S.20. Unit cell volume and lattice parameters of roubaultite under the effect of different external anisotropic pressures applied along the direction of minimum compressibility.

P (GPa)	Vol. (Å^3)	a (Å)	b (Å)	c (Å)	$\alpha\,(deg)$	$\beta\,(deg)$	$\gamma(deg)$
-0.0382	427.3044	6.9453	7.7712	7.9436	90.69	91.73	94.28
-0.0224	427.1795	6.9421	7.7807	7.9353	90.67	91.73	94.29
-0.0155	427.2545	6.9403	7.7860	7.9337	90.68	91.71	94.34
-0.0124	427.2554	6.9405	7.7887	7.9308	90.67	91.71	94.34
-0.0062	427.1576	6.9397	7.7903	7.9283	90.67	91.72	94.34
0.0007	427.0907	6.9385	7.7951	7.9235	90.65	91.72	94.34
0.0167	427.0634	6.9374	7.8010	7.9183	90.65	91.72	94.35
0.0225	427.0274	6.9369	7.8032	7.9161	90.64	91.71	94.36
0.0551	426.8094	6.9359	7.8064	7.9098	90.61	91.74	94.35

Table S.21. Calculated compressibilities ($k_V = -1/V \cdot (\partial V/\partial P)_P$) of roubaultite for different external anisotropic pressures applied along the direction of minimum compressibility.

P (GPa)	$k_V(TPa^{-1})$	**P (GPa)**	$k_V(TPa^{-1})$
-0.035	19.633	-0.015	-12.634
-0.030	27.453	-0.010	37.671
-0.025	11.788	-0.005	29.589
-0.020	-23.897	0.000	14.084
-0.0173	-33.958	0.005	3.066

Table S.22. Unit cell volume and lattice parameters of fontanite under the effect of different external anisotropic pressures applied along the direction of minimum compressibility.

P (GPa)	Vol. (Å^3)	a (Å)	b (Å)	c (Å)	$\alpha\,(deg)$	$\beta\,(deg)$	$\gamma(deg)$
-0.0806	1834.8349	6.9427	17.4221	15.1699	90.03	89.53	89.88
-0.0150	1828.9763	6.9449	17.4806	15.0661	90.00	89.53	89.97
-0.0031	1827.3408	6.9474	17.4875	15.0412	90.01	89.53	89.96
0.0018	1842.9043	6.9644	17.4162	15.1947	89.99	89.36	89.91
0.0191	1842.3470	6.9655	17.4199	15.1844	89.99	89.36	89.91
0.0352	1842.0804	6.9655	17.4206	15.1816	89.99	89.36	89.91
0.0370	1842.0410	6.9655	17.4209	15.1811	89.99	89.37	89.91
0.0661	1841.3159	6.9664	17.4217	15.1725	89.99	89.36	89.91
0.0671	1841.3050	6.9664	17.4219	15.1722	89.99	89.36	89.91
0.0786	1841.0487	6.9666	17.4225	15.1692	89.99	89.36	89.91

Table S.23. Calculated compressibilities ($k_V = -1/V \cdot (\partial V/\partial P)_P$) of fontanite under the effect of different external anisotropic pressures applied along the direction of minimum compressibility.

P (GPa)	$k_V(TPa^{-1})$	P (GPa)	$k_V(TPa^{-1})$	P (GPa)	$k_V(TPa^{-1})$
-0.050	47.237	-0.010	74.528	0.022	10.611
-0.045	50.638	-0.005	77.955	0.027	9.073
-0.040	54.042	-0.003	79.327	0.032	8.275
-0.035	57.448	0.000	-1431.168	0.037	8.217
-0.030	60.858	0.002	24.161	0.042	8.900
-0.025	64.270	0.007	19.664	0.050	11.533
-0.020	67.686	0.012	15.907	-	-
-0.015	71.106	0.017	12.889	-	-

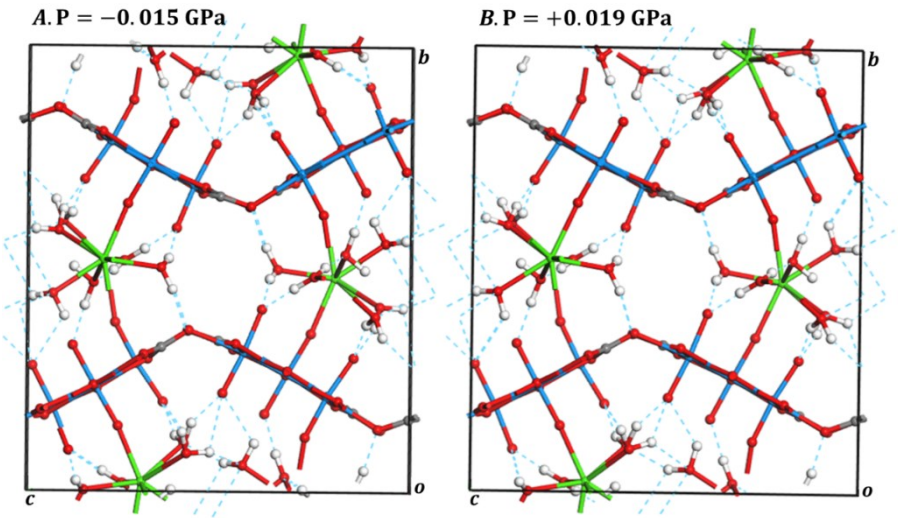

Fig. S.9. Views of the computed unit cell of fontanite from [100] under the effect of two different anisotropic pressures applied along the direction of minimum compressibility: (A) P = − 0.015 GPa; (B) P =+ 0.019 GPa.

Table S.24. Unit cell volume and lattice parameters of sharpite under the effect of different external isotropic pressures.

P (GPa)	Vol. ($Å^3$)	a (Å)	b (Å)	c (Å)
-0.9960	1814.4864	5.0328	15.9731	22.5710
-0.7413	1784.9822	5.0229	15.8416	22.4328
-0.5158	1765.9490	5.0162	15.7738	22.3184
-0.2479	1749.0213	4.9932	15.7664	22.2170
0.0017	1735.2999	4.9836	15.7296	22.1367
0.1248	1729.7926	4.9857	15.6946	22.1062
0.2554	1723.8019	5.0018	15.6166	22.0684
0.4997	1710.4719	4.9961	15.5514	22.0148
0.7436	1694.9453	4.9895	15.4792	21.9457
0.9886	1677.9576	4.9816	15.3834	21.8956

Table S.25. Calculated compressibilities ($k_a = -1/a \cdot (\partial a/\partial P)_P$) of sharpite for different external isotropic pressures.

P (GPa)	$k_V(TPa^{-1})$	P (GPa)	$k_V(TPa^{-1})$	P (GPa)	$k_V(TPa^{-1})$
-0.450	18.166	-0.100	6.450	0.200	-20.213
-0.400	18.627	-0.050	3.998	0.250	-8.038
-0.350	17.918	0.000	1.371	0.300	0.107
-0.300	16.300	0.050	-1.925	0.350	5.069
-0.250	14.075	0.100	-6.697	0.400	7.607
-0.200	11.547	0.150	-14.111	0.450	8.392
-0.150	8.959	0.185	-22.322	-	-

References

1 D. Ginderow, F. Cesbron, Structure de la roubaultite, $Cu_2(UO_2)_3(CO_3)_2O_2(OH)_2{\cdot}4\,H_2O$, *Acta Crystallogr. C*, 1985, **41**, 654.

2 K. A. Hughes, P. C. Burns, A new uranyl carbonate sheet in the crystal structure of fontanite, $Ca[(UO_2)_3(CO_3)_2O_2]{\cdot}(H_2O)_6$, *Am. Mineral*., 2003, **88**, 962.

3 J. Plášil, A unique structure of uranyl-carbonate mineral sharpite: A derivative of the rutherfordine topology, *Z. Kristallogr*., 2018, **233**, 579.

4 J. Plášil, L. Palatinus, J. Rohlíček, L. Houdková, M. Klementová, V. Goliáš, P. Škácha, Crystal structure of lead uranyl carbonate mineral widenmannite: Precession electron-diffraction and synchrotron powder-diffraction study, *Am. Mineral*., 2014, **99**, 276.

5 J. Plášil, K. Fejfarová, R. Skála, R. Škoda, N. Meisser, J. Hloušek, I. Císařová, M. Dušek, F. Veselovský, J. Čejka, J. Sejkora, P. Ondruš, The crystal chemistry of the uranyl carbonate mineral grimselite, $(K,Na)_3Na\left[(UO_2)(CO_3)_3\right]{\cdot}(H_2O)$, from Jáchymov, Czech Republic, *Mineral. Mag*., 2012, **76**, 443.

6 J. Plášil, K. Fejfarová, M. Dušek, R. Škoda, J. Rohlíček, Revision of the symmetry and the crystal structure of čejkaite, $Na_4[(UO_2)(CO_3)_3$, *Am. Mineral*., 2013, **98**, 549.