

The crystal structure of moctezumite $[\text{PbUO}_2](\text{TeO}_3)_2$

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ABSTRACT

Moctezumite, $[\text{PbUO}_2](\text{TeO}_3)_2$, is monoclinic, $P2_1/c$, with $a = 7.813(5)$, $b = 7.061(2)$, $c = 13.775(4)$ Å, $\beta = 93.71(2)^\circ$, and $Z = 4$. Its structure has been refined to an unweighted residual of 0.041 using 1440 reflections. Moctezumite possesses a layer structure parallel to (001). In each layer, U-coordination pentagonal bipyramids form zigzag chains parallel to **b** by sharing edges. The U chains alternate with chains of Pb edge-sharing monocapped trigonal-prism coordination polyhedra. The U-Pb layers stacked along **c*** are connected by Te_2O_6 units. Each Te_2O_6 unit is composed of $\text{TeO}_3\psi$ and $\text{TeO}_4\psi$ ($\psi = \text{lone pair}$), which share an O atom. The structure of moctezumite consists of slightly modified schmitterite-like (UTeO_5) winged U polyhedral chains separated by Pb polyhedral chains.

INTRODUCTION

Moctezumite was described and named by Gaines (1965). It was found at the Moctezuma mine near the town of Moctezuma, in north-central Sonora, Mexico. Compositionally, moctezumite is lead uranyl tellurite. It occurs as small, bright to dark orange, bladed crystals, usually multiple, in hydrothermal quartz veins in the oxidized zone of a Te-Au deposit. From a wet chemical analysis, Gaines (1965) assigned the formula $\text{PbO} \cdot \text{UO}_3 \cdot 2\text{TeO}_2$, and measured a density of 5.73 g/cm^3 using a pycnometer. Because of the difficulty in getting a suitable single crystal of moctezumite, attempts were made to hydrothermally synthesize moctezumite. Brandstätter (1981a) obtained small, orange, needle-shaped single crystals of $\text{Pb}_2[\text{UO}_2][\text{TeO}_3]_3$, which is not the same as moctezumite, and determined its structure. R.V.G. succeeded in synthesizing moctezumite, but the material was very fine grained with no usable crystals. As of mid-1992, only two other Te minerals containing U have been reported, both also present at the Moctezuma mine. These minerals are cliffordite (Gaines, 1969) and schmitterite (Gaines, 1971). Galy and Meunier (1971) determined the structure of synthetic cliffordite, UTe_3O_5 , and Meunier and Galy (1973) determined the structure of synthetic schmitterite, UTeO_5 .

EXPERIMENTAL METHODS

Upon recent reexamination of original material from the Moctezuma mine, a crystal fragment was isolated that

appeared to be large enough for X-ray analysis. The fragment was broken into some smaller fragments for X-ray analysis. Prior to initiating data measurement, Weissenberg photographs of the crystal were taken, which indicated monoclinic symmetry. A transparent, light orange, anhedral crystal was mounted on a rotating anode CAD4 diffractometer with graphite monochromated (graphite $2\theta = 12.2^\circ$) $\text{MoK}\alpha$ radiation (0.71069 Å). Cell parameters and an orientation matrix for data measurement were obtained from a least-squares refinement using the setting angles of 24 centered reflections in the range $11.43 < 2\theta < 23.83^\circ$. Based on systematic absences of $h0l$, $l = 2n$, and $k = 2n$ based on systematic absences of $0k0$; the space group was determined to be $P2_1/c$. The cell parameters and the space group confirmed the earlier determined parameters (Gaines, 1965).

Three peaks were selected and used as intensity standards for monitoring the measurements at 7200-s intervals. The intensities of the standard peaks varied $< 2\%$ during data collection. The same standard peaks were used to check the orientation of the crystal after every 200 intensity measurements. If an angular error of $> 0.10^\circ$ was observed, a new orientation matrix was automatically established by recentering the original 24 diffraction maxima. The intensities were corrected for Lorentz, polarization, and absorption factors (the latter using ψ -scans). Experimental details are summarized in Table 1.

STRUCTURE DETERMINATION

Solution of the crystal structure was straightforward with direct methods (Hall and Stewart, 1989). The heavy

† Deceased August, 1992.

TABLE 1. Crystal and experimental data of moctezumite

Crystal-cell data	
<i>a</i> (Å)	7.813(5)
<i>b</i> (Å)	7.061(2)
<i>c</i> (Å)	13.775(4)
β (°)	93.71(2)
<i>V</i> (Å ³)	758.3(6)
FW	828.42
<i>F</i> (000)	1368.00
Space group	<i>P</i> 2 ₁ / <i>c</i>
<i>Z</i>	4
Formula	[PbUO ₂] ₂ (TeO ₃) ₂
<i>D</i> _{calc} (g/cm ³)	7.256
μ (mm ⁻¹)	37.61
Intensity measurements	
Crystal size (mm)	0.06 × 0.08 × 0.10
Diffractometer	Enraf-Nonius Cad4
Monochromator	Graphite
Radiation	MoK α_1 (cell) (= 0.70926 Å) MoK α (data) (= 0.71069 Å)
Scan type	θ -2 θ
2 θ range	0.5–55.8°
Max. counting time (s)	60
<i>hkl</i> ranges	$\bar{1}$ 0–10, 0–9, 0–18
No. of refl. measured	3507
No. of unique refl.	1832
No. of refl. with $F_o > 3.0\sigma(F_o)$	1440
Absorption corrections were made using ψ -scans.	
Min. and max. transmission factors	0.21 and 0.99
Refinement of the structure	
Residuals:	
For significant reflections	<i>RF</i> 0.041 <i>R_w</i> 0.049 GoF 3.84
For all reflections	<i>RF</i> 0.059 <i>R_w</i> 0.064
where	$RF = \text{Sum}(F_o - F_c) / \text{Sum}(F_o)$ $R_w = \text{Sqrt}\{\text{Sum}[w(F_o - F_c)^2] / \text{Sum}(wF_o^2)\}$ $GoF = \text{Sqrt}\{\text{Sum}[w(F_o - F_c)^2] / (\text{no. of refls.} - \text{no. of params.})\}$

Note: The last least-squares cycle was calculated with 12 atoms, 70 parameters, and 1440 reflections. Unit weights were used. The maximum shift/ σ ratio was 0.000. In the last difference synthesis, the largest minimum was -3.890 e/Å³, and the highest peak was 4.780 e/Å³. Secondary ext. coeff. = 0.000265; $\sigma = 0.000601$.

U, Pb, and Te atoms were located. A difference-Fourier map, phased with the above mentioned atoms, revealed positions of all O atoms in the asymmetric unit. Positional and thermal parameters (initially isotropic) were refined using a block-diagonal least-squares program (NRCVAX, Gabe et al., 1989). Attempts were made to refine all the atoms with anisotropic displacement parameters using the full-matrix least-squares method. All O atoms showed some nonpositive displacement parameters, probably caused by high absorption of X-rays by the crystal. The shape of the crystal used for data measurement was such that analytical absorption corrections were not possible; hence ψ absorption corrections were applied. The final refinement was carried out with 70 variables, including positional parameters and anisotropic displacement factors for Pb, U, and Te atoms and positional parameters for O atoms. Isotropic displacement factors were used for O atoms. Full-matrix least-squares refinements were used during final cycles. The atomic scattering factors for Pb, U, Te, and O were taken from the *International Tables for X-ray Crystallography* (Ibers and Hamilton, 1974) and were corrected for anomalous

TABLE 2. Atomic parameters *x*, *y*, *z*, and *B*_{iso} (Å²) for moctezumite

	<i>x</i>	<i>y</i>	<i>z</i>	<i>B</i> _{iso}
U	0.0502(1)	0.1564(1)	0.80919(7)	0.71(4)
Pb	0.5642(1)	0.1241(1)	0.32911(7)	1.23(4)
Te1	0.3500(2)	0.1855(2)	0.0450(1)	0.79(5)
Te2	0.8158(2)	0.1669(2)	0.5806(1)	0.78(6)
O1	0.161(2)	0.856(3)	0.146(1)	1.4(3)*
O2	0.296(2)	0.447(3)	0.036(1)	1.4(3)*
O3	0.739(2)	0.825(3)	0.237(1)	1.6(3)*
O4	0.615(2)	0.346(3)	0.143(1)	1.5(3)*
O5	0.485(2)	0.820(3)	0.043(1)	1.4(3)*
O6	0.934(2)	0.526(3)	0.155(1)	1.6(4)*
O7	0.925(2)	0.166(3)	0.181(1)	1.5(3)*
O8	0.832(2)	0.884(3)	0.042(1)	1.9(4)*

Note: *B*_{iso} is the mean of the principal axes of the thermal ellipsoid. Estimated standard deviations refer to the last digit printed.

* These atoms were refined isotropically.

dispersion. The final unweighted *R* factor was 0.041, and the results of the refinement are shown in Table 1. Final atomic coordinates and isotropic thermal parameters are given in Table 2. Anisotropic thermal parameters are given in Table 3.¹ The observed and calculated structure factors are listed in Table 4.¹

The structural formula for moctezumite deduced from the refinement is [PbUO₂]₂(TeO₃)₂. The cell content and density differ from the earlier reported chemical data (Gaines, 1965). Instead of 3, *Z* is found to be 4. The density calculated from the data presented here is 7.256 g/cm³ as opposed to the value 5.73 g/cm³ measured by Gaines (1965). Given the difficulties described by Gaines (1965) in completely separating moctezumite for chemical analysis, it is possible that the measured density was compromised by the presence of small quantities of other phases.

STRUCTURE DESCRIPTION

Moctezumite possesses a layer structure. The U and Pb polyhedra are located in layers parallel to (001) (Fig. 1). In each layer, U pentagonal bipyramids form zigzag chains by sharing equatorial edges (O6-O7). In each chain U atoms are 3.95 Å apart, and the U-U-U angle is 126.89°. Similar chains have been reported in synthetic schmitterite, UTeO₅ (Meunier and Galy, 1973). The U polyhedral chains are parallel to the *b* axis and are connected to other U chains on both sides by zigzag chains of Pb polyhedra. The bladed nature of moctezumite crystals elongated parallel to *b* (Gaines, 1965) is an expression of the polyhedral chains in the structure. The U pentagonal bipyramid shares its axial vertices (O1 and O3) with the Pb coordination polyhedron. The Pb polyhedral chains are formed of edge-sharing monocapped trigonal prism coordination polyhedra. The zigzag nature of the U and Pb polyhedral chains is generated by screw axes parallel

¹ Copies of Tables 3 and 4 may be ordered as Document AM-93-533 from the Business Office, Mineralogical Society of America, 1130 Seventeenth Street NW, Suite 330, Washington, DC 20036, U.S.A. Please remit \$5.00 in advance for the microfiche.

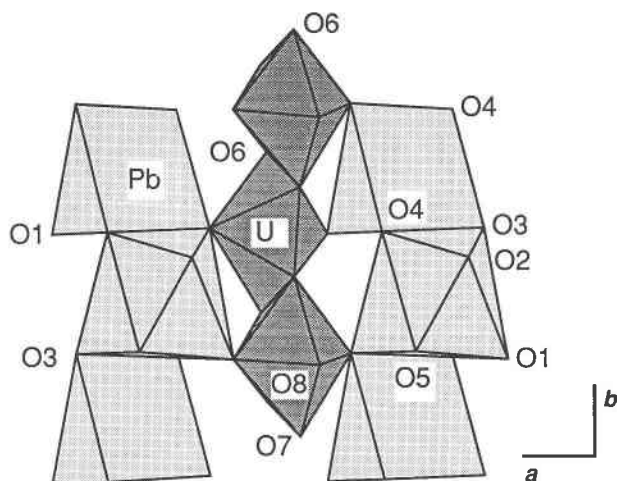


Fig. 1. The layer structure of moctezumite parallel to (001) (projected along c^*). The layers consist of alternating Pb and U polyhedral chains parallel to b . All figures were produced, in part, with the Atoms program (Shape Software, Kingston, Tennessee).

to b at $x = 0$ and $\frac{1}{2}$ and $Z = \frac{1}{4}$ and $\frac{3}{4}$. Neighboring U or Pb coordination polyhedra in a chain are rotated 180° about b relative to one another.

U-Pb layers stacked along c are connected to each other by Te coordination units. Figure 2 shows the U-Pb layers viewed end on (down b) and the Te-O units interspersed between the layers. Te1 and Te2 bridge the U and Pb chains in a layer and also bridge neighboring U-Pb layers along c . There are two U-Pb layers stacked along c in the unit cell, and they are related to each other by the inversion point at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$. Te1 and Te2 form a Te_2O_6 unit by sharing O2. The lone pair of electrons of Te1 and Te2 appear to be accommodated in the relatively open spaces between the U-Pb layers.

Table 5 shows bond valence sums for moctezumite. The sum for U is rather high at 6.71 vu. However, in synthetic cliffordite (Brandstätter, 1981b) the U sum is 6.60. Given that in moctezumite the U ligands O1 and O3 are overbonded by about the same amount as U, we feel that the r_o and B values for U reported by Brown and Altermatt (1985) may need reconsideration. The other cation sums are satisfactory.

DESCRIPTION OF THE POLYHEDRA

U polyhedron

There is only one U atom in the asymmetric unit. It forms an uranyl group with O1 and O3 in which O-U-O = 179° and the average U-O bond length is 1.80 Å (Table 6). U^{6+} is bonded to five other O atoms, and altogether the seven ligands form a distorted pentagonal bipyramid coordination polyhedron. The distances between U and the equatorial O atoms (O6b, O6c, O7c, O7d, and O8) range from 2.20 to 2.59 Å, the average being 2.36 Å. A least-squares plane through the equatorial atoms dem-

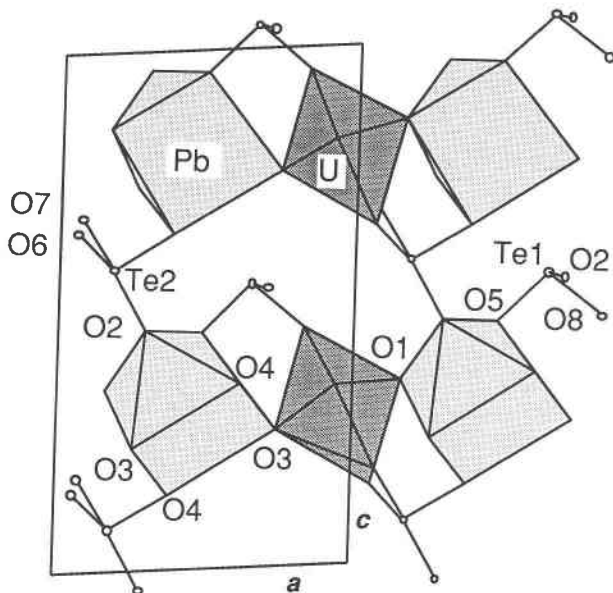


Fig. 2. The structure projected down the b axis (half of the b translation) showing the U-Pb layers viewed edge-on and their linkage by Te1 and Te2. Pb and U coordinations are shown as polyhedra and Te coordinations are shown by spokes.

onstrates that these atoms define a surface that is nearly planar. The distances of O6b, O6c, O7c, O7d, and O8 from this plane range from -0.04 to 0.08 Å, whereas atoms O1 and O3 (the axial ligands) are 1.81 and -1.83 Å away from this plane. The U atom lies on the equatorial plane. Similar U^{6+} coordinations have been reported in synthetic schmitterite (Meunier and Galy, 1973), synthetic $Pb_2[UO_2][TeO_3]_2$ (Brandstätter, 1981a), other U compounds (Pertlik, 1974; Brandenburg and Loopstra, 1973, 1978; Loopstra and Brandenburg, 1978), and the minerals kasolite, vandenbrandeite (Rosenzweig and Ryan, 1977a, 1977b), and sklodowskite (Ryan and Rosenzweig, 1977).

Pb mon capped trigonal prism

The one Pb atom in the asymmetric unit is in sevenfold coordination with O atoms. The coordination polyhe-

TABLE 5. Bond valence calculations for moctezumite

	U^{6+}	Pb^{2+}	$Te1^{4+}$	$Te2^{4+}$	Σ
O1	2.09	0.20			2.29
O2		0.42	1.24	0.34	2.00
O3	2.07	0.13			2.33
		0.13			
O4		0.39		1.44	1.91
		0.08			
O5		0.62	1.47		2.09
O6	0.55			1.19	1.98
	0.24				
O7	0.56			1.00	2.07
	0.51				
O8	0.69		1.34		2.03
Σ	6.71	1.97	4.05	3.97	

Note: Calculations by the method of Brown and Altermatt (1985).

TABLE 6. Bond distances (Å) and angles (°) of moctezumite

U pentagonal dipyramid		Pb monocapped trigonal prism	
U-O1	1.80(2)	Pb-O5	2.28(2)
-O3	1.80(2)	-O2	2.43(2)
-O8	2.20(2)	-O4e	2.45(2)
-O7d	2.28(2)	-O1	2.70(2)
-O6b	2.29(2)	-O3f	2.85(2)
-O7c	2.32(2)	-O3e	2.86(2)
-O6c	2.59(2)	-O4g	3.04(2)
Average	2.18	Average	2.66
Te1 TeO ₃ pyramid		Te2 TeO ₄ tetrahedron	
Te1-O5	1.83(2)	Te2-O4	1.84(2)
-O8	1.86(2)	-O6	1.91(2)
-O2	1.89(2)	-O7	1.97(2)
		-O2	2.36(2)
Average	1.86	Average	2.02

Note: Standard deviations of the last digit are in parentheses. Symmetry codes: b = 1 - x, 1 - y, 1 - z; c = -1 + x, 1/2 - y, 1/2 + z; d = 1 - x, -y, 1 - z; e = -x, -1/2 + y, 1/2 - z; f = -1 + x, -1 + y, z; g = -1 + x, y, z.

dron can be described best as a distorted monocapped trigonal prism. Pb-O distances range from 2.28 to 3.04 Å, the average being 2.66 Å (Table 6). The two non-capped prism faces of the Pb coordination polyhedron are nearly flat. The deviations from the least-squares surfaces through O3e, O3f, O4e, and O4g, and O1, O3e, O4g, and O5 range from -0.06 to 0.06 and -0.03 to 0.03 Å, respectively. O2 forms the capping vertex on the third side of the prism. Pb in seven- and eightfold-coordination is present in Pb₂[UO₂][TeO₃]₃ (Brandstätter, 1981a). Brandstätter (1981a) observed that the O atoms around Pb in Pb₂[UO₂][TeO₃]₃ show a strong tendency to a one-sided coordination, owing to the presence of a lone pair of 6s electrons. Such interaction between lone pairs and bond pairs is not evident around Pb in moctezumite.

Te polyhedra

There are two Te atoms in the asymmetric unit of moctezumite. Te1 forms three bonds with O atoms, and Te2 forms three short bonds and one longer bond with O atoms. Te1-O and Te2-O distances range from 1.83 to 1.89 and 1.84 to 2.36 Å, respectively. We consider that Te1 does not form bonds with O4 and O5h (distances 2.66 and 2.78 Å), because the bond valence contributions would each be <3.5% of the sum, and Te1 is saturated without them (Table 5). O5 is not considered to bond to Te2 because of its very small bond valence (0.05 vu, bond distance 3.02 Å).

The dispositions of the Te atoms relative to their coordinating O atoms is interesting. In the case of Te1, the coordinating O atoms are O2, O5, and O8, and they are all located to one side of Te1. Te2 is coordinated by O2, O4, O6, and O7 and is situated within the polyhedral volume bounded by these atoms but is located very nearly on one edge close to its midpoint. The one-sided disposition of the coordinating O atoms of each Te atom reflects the interaction between bond pairs and the lone

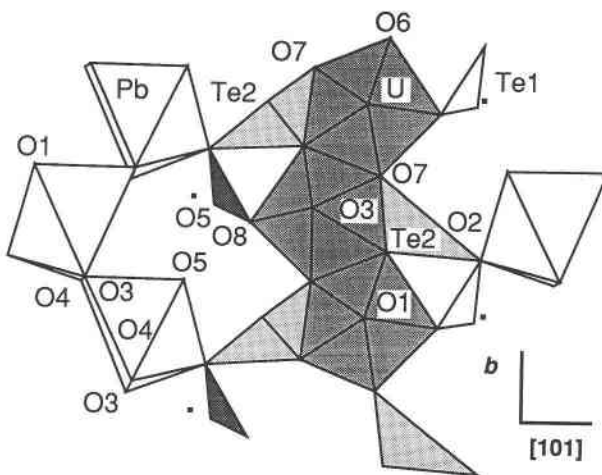


Fig. 3. A view of a portion of the structure parallel to (10T). Darker shaded Te1 coordinating O atom planes plunge down into the page, whereas unshaded ones project up out of the page. The locations of the Te1 atoms relative to their ligands are indicated by dots. In this orientation, the relationship to the U-Te chain structure of schmitterite (U-TeO₃) is evident (see text).

pair. If the Te lone pair of electrons is considered as a member of the Te coordination polyhedron, then Te1 is tetrahedrally coordinated, and Te2 is in trigonal bipyramidal coordination. In effect, the lone pair of Te2 forms one of the equatorial vertices of a trigonal bipyramid. These coordination geometries for Te⁴⁺ have been noted in many other structures and are well discussed in the literature (e.g., Galy et al., 1975).

Te-O coordinations like the ones in moctezumite have been reported for synthetic cliffordite (Brandstätter, 1981b), uranyl tellurite (Loopstra and Brandenburg, 1978), and several other structures.

STRUCTURE COMPARISON

Moctezumite is the only mineral reported to date containing essential Pb, U, and Te. However, the structure of moctezumite may be compared with those of synthetic Pb₂[UO₂][TeO₃]₃ and synthetic schmitterite. Synthetic Pb₂[UO₂][TeO₃]₃ (Brandstätter, 1981a) contains three Te atoms, two Pb atoms, and one U atom in the asymmetric unit. The structure contains (1) one Pb in sevenfold and the other in eightfold coordination with O atoms, (2) two TeO₃ pyramids and the third Te in 3 + 1 coordination, and (3) U in sevenfold coordination with O atoms. The structure is described as having sheets parallel to (010) composed of U and Te polyhedra. The sheetlike {[UO₂][TeO₃]}_n⁴ⁿ⁻ units are held together along b by Pb polyhedral chains parallel to c. In moctezumite, U-Te layers are found to occur parallel to (10T). However, these layers are made up of strips, each of which consists of a U pentagonal bipyramidal chain bearing Te coordination units on either side as wings. The strips are held together by Pb polyhedral chains (parallel to b) sandwiched between the U-Te layers.

Synthetic schmitterite (Meunier and Galy, 1973) contains no Pb, but the dispositions of U and Te in it may be fruitfully compared with those in moctezumite. In schmitterite, U pentagonal bipyramids form chains that are very similar to the U polyhedral chains of moctezumite. The U chains are crosslinked in the (010) plane by $\text{TeO}_4\psi$ units (where ψ is the lone pair of electrons). These units are essentially the same as the Te_2 coordination environment in moctezumite. However, the U-Te equatorial edge-sharing topologies are slightly different in the two structures. Nevertheless, when projected on the (10 $\bar{1}$) plane (Fig. 3), the structure of moctezumite can be seen to consist of schmitterite-like U-Te strips parallel to **b**, separated from one another by Pb chains.

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