A NEW URANYL SHEET IN K₅[(UO₂)₁₀O₈(OH)₉](H₂O):
NEW INSIGHT INTO SHEET ANION-TOPOLOGIES

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ABSTRACT

The structure of K₅[(UO₂)₁₀O₈(OH)₉](H₂O), Z = 4, monoclinic, space group Pn, a 13.179(2), b 20.895(4), c 13.431(3) Å, β 106.316(3)°, has been solved and refined to R = 7.8% and a goodness-of-fit of 0.82 for 4349 unique observed (F₀ ≥ 4σF) reflections collected for a synthesized crystal using MoKα X-radiation and a CCD detector. The structure is dominated by previously unknown sheets of edge- and vertex-sharing uranyl polyhedra parallel to (100), with K cations and H₂O groups in the interlayer. The sheets contain 20 symmetrically distinct U⁶⁺ cations, each of which is associated with an approximately linear (UO₂)²⁺ uranyl ion (Ur), and is coordinated by four or five additional anions at the equatorial positions of square (2 polyhedra) and pentagonal (18 polyhedra) bipyramids, respectively. As is the case with other sheets of uranyl polyhedra, the anion topology of the sheet can be generated using a chain-stacking sequence; however, the sequence is unique and for the first time involves the occurrence of P chains flanked by U or D chains of opposite directional sense. This example serves to further demonstrate the structural complexity that is possible in sheets of uranyl polyhedra.

Keywords: uranium, uranyl oxide hydrate of potassium, synthetic, anion topology, crystal structure.

INTRODUCTION

There are ~200 uranyl minerals (Mandarino 1999) that are important for understanding the genesis of U deposits (Frondel 1958, Finch & Ewing 1992). Uranyl minerals are significant in several areas of environmental concern. They are present in the mill tailings that result from the mining of U, and are precipitated in soils that are contaminated with actinide elements. They will be the dominant products of alteration of spent nuclear fuel in a geological repository if the conditions are oxidizing and the fuel comes in contact with water, such as is likely to occur at the proposed repository at Yucca Mountain, Nevada (Finn et al. 1996, Wronkiewicz et al. 1996). An understanding of the complex crystal chemistry of these minerals is essential to the prediction of their impact on the release of radionuclides from nuclear waste under repository conditions.
The paragenesis of uranyl minerals should be related to the energetics and stabilities of the underlying crystal structures. However, the links will only be revealed following a substantial improvement of our knowledge of the crystal chemistry of uranyl minerals, which lags well behind that of many other mineral groups (despite their obvious importance). The structures have been determined and refined for only ~60 uranyl minerals, less than one-third of the known species. This is due to severe experimental difficulties; uranyl minerals are extreme absorbers of X-rays, many do not form crystals of a size suitable for structure analysis using conventional techniques, and others dehydrate, causing deterioration of the crystals. The recent introduction of CCD-based X-ray detectors to mineralogical studies (Burns 1998a) has resulted in significant advances in our understanding of the structures of uranyl minerals (Burns 1997, 1998b, c, d, 1999a, 2000a, b, Burns & Finch 1999, Burns & Hanchar 1999, Hill & Burns 1999, Burns & Hill 2000). Surprising discoveries include the presence of $\text{U}^{5+}$ in wyartite (Burns & Finch 1999), uranyl phosphate chains in parsonsite (Burns 2000b), and extraordinarily complex sheets of uranyl polyhedra in vandendriesscheite (Burns 1997) and wölsendorfite (Burns 1999a). Following the discovery of the wölsendorfite sheet with a primitive repeat of 56 Å, Burns (1999a) noted that such complex sheets contain components of two or more simpler sheets observed in other minerals, suggesting that many new sheets of uranyl polyhedra may await discovery. A substantial portion of the structural energetics of uranyl minerals must depend upon the connectivity of the sheets of polyhedra. The details of the sheet anion-topologies may therefore be a key to understanding and predicting the thermodynamics of these phases.

We have undertaken studies of synthetic ($<200^\circ$C) uranyl phases to obtain new insight into the structures, stabilities, and chemistries of uranyl minerals. Crystals of a K uranyl oxide hydrate (hereafter designated $\text{KUOH}$) that contains a new sheet of uranyl polyhedra have been obtained, and the details of the structure are presented herein.

**EXPERIMENTAL METHODS**

**Synthesis of crystals**

Amber-yellow crystals of $\text{KUOH}$ were obtained from synthesis experiments intended to give boltwoodite, modified from the methods of Vochten et al. (1997). A solution containing 0.1M uranium oxynitrate (Alfa Æsar), $\text{UO}_2\left(\text{NO}_3\right)_2\cdot6\text{H}_2\text{O}$, and 0.1M KCl (Fisher (1997). A solution containing 0.01M uranium oxynitrate was adjusted to ~12 using 5M KOH (Fisher Scientific). One gram of natural quartz crystals was added to the solution in a Teflon-lined Parr bomb. The reactants were held at 225°C for 72 h, and the products were allowed to cool slowly. The resulting crystals were separated from the solution by filtration and were washed three times using ultrapure water. The crystals consisted of ~95% boltwoodite and ~5% of $\text{KUOH}$.

**X-ray diffraction**

An acicular crystal with sharp extinction and uniform optical properties was selected for the X-ray study. The crystal was mounted on a Bruker PLATFORM 3-circle goniometer equipped with a 1K SMART CCD (charge-coupled device) detector and a crystal-to-detector distance of 5 cm (Burns 1998a).

More than 75% of the sphere of data was collected to ~57° 20° using monochromatic MoKα X-radiation and frame widths of 0.3° in $\omega$, with 180 s used to acquire each frame. The final unit-cell dimensions (Table 1) were refined on the basis of 2785 reflections using least-squares techniques. Data were collected in approximately 95 hours; comparison of the intensities of equivalent reflections collected at different times during the data collection showed no evidence of significant decay. The three-dimensional data were reduced and corrected for Lorentz, polarization, and background effects using the Bruker program SAINT. An empirical absorption-correction was done using the program SADABS (G. Sheldrick, unpubl. computer program) on the basis of the intensities of equivalent reflections. A total of 32,837 reflections was collected; the merging of equivalent reflections gave 16,067 unique reflections, with 4349 classed as observed ($F_o \geq 4\sigma F_o$).

**STRUCTURE SOLUTION AND REFINEMENT**

Scattering curves for neutral atoms, together with anomalous dispersion corrections, were taken from *International Tables for X-Ray Crystallography, Vol. IV* (Ibers & Hamilton 1974). The Bruker SHELXTL Version 5 system of programs was used for the refinement of the crystal structure. Reflection statistics and systematic absences indicated space groups $Pn$ or $P21n$. Attempts to solve the structure in the centrosymmetric space group failed, and space group $Pn$ was verified by the successful solu-

| $a$ (Å) | 13.178(2) |
| $b$ (Å) | 20.895(4) |
| $c$ (Å) | 13.431(3) |
| $\alpha$ | 106.316(3) |
| $\beta$ | 114.95(9) |
| $\gamma$ | 114.95(9) |

| Space group | $Pn$ |
| $F(000)$ | 5320 |
| $\mu$ (mm$^{-1}$) | 46.1 |
| $D_{calc}$ (g/cm$^3$) | 5.979 |

Unit-cell contents: $4\left[K\left(UO_2\right)_3\left(O_3\right)_2\left(OH\right)_2\right]\left(H_2O\right)$

$R = \frac{\sum||F_o|| - |F_c||}{\sum|F_o|}$

$S = \frac{\sum[|F_o|^2 - |F_c|^2]^2/|F_o|^2}{\sum|F_o|^2}$, for $m$ observations and $n$ parameters.
tion of the structure by direct methods. The initial model included the positions of the U atoms, with K cations and anions located in difference-Fourier maps after least-squares refinement of the model. The final model involved refinement of the atomic positional parameters, isotropic-displacement parameters, and a weighting scheme for the structure factors. Refinement of this very large structure was difficult owing to weak data and pseudosymmetry. Some of the uranyl ion $U^+$–O bond-lengths refined to values outside the range in well-refined structures; the “soft” constraint that these bond lengths be ~1.8 Å was imposed during the final cycles of refinement, and did not change the agreement index ($R$). Two overall displacement parameters were refined for the anions, one corresponding to O and the other to OH and H$_2$O groups. Refinement of anisotropic-displacement parameters for the cations only reduced $R$ by ~0.5%, and led to instability of the refinement. The final $R$ was 7.8% for the isotropic model for $349$ unique observed ($F_o \geq 4\sigma F$) reflections, and the goodness-of-fit ($S$) was 0.82. The final atom parameters are given in Table 2, and selected mean interatomic distances and angles are given in Table 3. Calculated and observed structure-factors are available from the Depository of Unpublished Data, CISTI, National Research Council, Ottawa, Ontario K1A 0S2, Canada.

### Table 2: Atom Parameters for KUOH

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<th>x</th>
<th>y</th>
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<td>0.338(2)</td>
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<td>(O4)</td>
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<td>0.275(4)</td>
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<td>(O5)</td>
<td>0.149(3)</td>
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<tr>
<td>(O6)</td>
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<td>0.137(4)</td>
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### Table 2 (continued): Atom Parameters for KUOH

<table>
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$\tilde{u}_{eq} = \frac{1}{3} \sum_{i=1}^{3} u_{ij} \tilde{R}^{ij} x^2$
DESCRIPTION OF THE STRUCTURE

Projection of the structure along [100] (Fig. 1) reveals the presence of sheets of uranyl polyhedra parallel to (100), with K cations and H2O groups located in the interlayer. This is in accord with other uranyl oxide hydrate structures, each of which contain sheets of polyhedra of higher bond-valence, with lower-valence cations and H2O groups located in interlayer positions. This structural theme also dominates other groups of uranyl structures, including uranyl silicates, phosphates, vanadates, arsenates, and molybdates (Burns et al. 1996).

Formula of KUOH

All atoms in the structure are on general positions in the space group Pn (Table 2). Bond-valence sums calculated using parameters for U–O bonds (Burns et al. 1997b) and K–O bonds (Brese & O’Keeffe 1991) permitted the distinction of O, (OH)– and H2O. The formula of the crystal studied is therefore K5[(UO2)10O8(H2O)], Z = 4.

Cation polyhedra

Twenty symmetrically distinct U6+ cations occur in the structure. Each is strongly bonded to two O atoms, resulting in approximately linear (UO2)2+ uranyl ions (Ur), as is almost invariably the case in uranyl phases (Burns et al. 1997b). The mean U–OUr bond lengths are all ~1.8 Å, as required by the constraints imposed on the refinement, with OUr–U–OUr bond angles ranging between 162(2) and 179(3)°. Eighteen of the uranyl ions...
are coordinated by five anions arranged at the equatorial vertices of pentagonal bipyramids capped with the $O_{Ur}$ atoms. The pentagonal bipyramid is the most common $U^{6+}$ coordination polyhedron in uranyl minerals (Evans 1963, Burns et al. 1997b). The mean U–O$_{eq}$ (equatorial) bond lengths range from 2.33 to 2.44 Å, in accord with the mean value of 2.37(9) Å for U–O$_{eq}$ bonds in many $Ur\phi_4$ polyhedra in well-refined structures (Burns et al. 1997b). The equatorial anions of twelve of these polyhedra are comprised of two O atoms and three (OH)$^-$ groups, whereas the other six contain three O atoms and two (OH)$^-$ groups. Two uranyl ions, U(6) and U(10), are coordinated by four anions arranged at the vertices of strongly distorted square bipyramids capped by the $O_{Ur}$ atoms. This $U^{6+}$ polyhedron is common in uranyl phosphates and arsenates that contain sheets based upon the autunite anion-topology (Burns et al. 1996), but is not common in sheets together with uranyl pentagonal bipyramids. Exceptions are curite (Taylor et al. 1981), its synthetic Sr analog (Burns & Hill 2000), and sayrite (Piret et al. 1983). The $Ur\phi_4$ polyhedra in $KUOH$ are strongly distorted (Fig. 2), and are similar to the polyhedra in the curite sheet (Taylor et al. 1981, Burns & Hill 2000). The equatorial anions in each correspond to three O and one (OH)$^-$, with the mean U–O$_{eq}$ bond lengths being 2.28 and 2.34 Å, in agreement with the mean U–O$_{eq}$ bond length of 2.28(5) in numerous $Ur\phi_4$ polyhedra in well-refined structures (Burns et al. 1996).

The structure contains ten symmetrically distinct K cations, each of which is located in the interlayer. Several of the $K\phi_4$ polyhedra are surrounded by ligands at distances in the range 3.0 to 3.4 Å, making description of the coordination polyhedra somewhat subjective. All K cations are coordinated by at least six ligands with bond lengths less than 3.1 Å, and some have as many as ten ligands within 3.4 Å. Coordinating ligands correspond to $O_{Ur}$ anions, O and (OH)$^-$ anions located at the equatorial positions of uranyl polyhedra within the sheets, and H$_2$O groups in the interlayer. The $K\phi_4$ polyhedra share edges and faces, forming double-wide chains that are parallel to [010] (Fig. 3).

Sheets of uranyl polyhedra

Uranyl polyhedra polymerize by edge- and vertex-sharing, forming complex sheets parallel to (100) (Fig. 2), with the uranyl ions oriented roughly perpendicular to the plane of the sheet. The sheet, with a primitive repeat-distance of 20.9 Å, is more complex than
most sheets of uranyl polyhedra, except for the vandendriesscheite sheet (41.4 Å) and the wölsendorfite sheet (56.0 Å). The sheet is topologically unique amongst minerals and synthetic uranyl phases; thus it provides additional insights into the complex connectivities of such sheets.

**Sheet Anion-Topologies in Uranyl Oxide Hydrates**

The sheet anion-topology approach for comparing sheets of uranyl polyhedra and other polyhedra of higher bond-valence was introduced by Burns *et al.* (1996). It is not unusual for substantially different sheets to correspond to the same underlying topological arrangement of anions. Miller *et al.* (1996) developed the sheet anion-topology approach for uranyl oxide hydrates, and introduced representations of the topologies as chain-stacking sequences. With the exception of the curite anion-topology, only four chains of polygons are required to construct each of the anion topologies that correspond to sheets that contain only uranyl polyhedra (square, pentagonal, or hexagonal bipyramids); these are shown in Figure 4a. The P chain is composed of edge-sharing pentagons, the R chain contains rhombs, and the H chain contains edge-sharing hexagons. The arrowhead chains (U and D), which have a directional aspect, contain both pentagons and triangles, arranged such that each triangle shares an edge with a pentagon, and the opposite corner with another pentagon in the chain.

Generation of anion topologies using chain-stacking sequences is an efficient way of understanding the relationships among the complex sheets recently found in several minerals. This approach may be a key to understanding the links between complex structures, such as those of vandendriesscheite and wölsendorfite, and mineral paragenesis. Here we provide an overview of the different anion-topologies and corresponding chain-stacking sequences for sheets in uranyl oxide hydrate minerals and synthetic compounds, and restrict our discussion to sheets that contain only uranyl polyhedra. Burns (1999d) extended this approach to various sheets that contain other cation polyhedra of higher bond-valence in addition to uranyl.

**Protasite (α-U$_3$O$_8$) anion-topology**

The protasite anion-topology (Fig. 5a) is the basis of sheets that occur in protasite, becquerelite, billietite (Pagoaga *et al.* 1987), richetite (Burns 1998b), com-
preignacite (Burns 1998c), agrinierite (Cahill & Burns 2000) and masuyite (Burns & Hanchar 1999), as well as a synthetic Cs uranyl oxide hydrate (Hill & Burns 1999). As such, it is apparent that sheets based upon this topology are compatible with a range of interlayer configurations involving Ba, Ca, Pb, K and Cs. This simple anion-topology contains only triangles and pentagons. Only P and D chains are required to develop the protasite anion-topology, with the repeat sequence PDPD...

Fourmarierite anion-topology

Sheets based upon the fourmarierite anion-topology (Fig. 5b) occur in fourmarierite (Piret 1985) and schoepite (Finch et al. 1996). In schoepite, the sheets are neutral, with H₂O groups being the only constituents of the interlayer. In fourmarierite, the sheets have a different distribution of O and (OH)⁻, resulting in a charged sheet that is balanced by Pb²⁺ in the interlayer. The anion topology can be obtained with a chain-stacking sequence involving P, D and U chains, with the sequence DUPUDPU...

Vandendriesscheite anion-topology

The vandendriesscheite anion-topology (Fig. 5c) is exceptionally complex, with a primitive repeat of 41.4 Å (Burns 1997). Only vandendriesscheite contains a sheet based upon this topology, which can be constructed using P, U and D chains in the sequence PDUPUPUPUDPDPDPUPUPUPU... It contains sections that are identical to the PDPD... (or PUPU...) repeats of the protasite anion-topology, with the join between such sections involving the DU sequence of the fourmarierite anion-topology. Thus, the vandendriesscheite anion-topology is a structural intermediate between these two simpler anion-topologies.

Sayrite anion-topology

Sayrite (Piret et al. 1983) is the only mineral that contains sheets based upon the sayrite anion-topology, which involves U, D, P, and R chains (Fig. 5d). The chains are arranged such that each P chain is flanked by two arrowhead chains with the same sense (direction), giving UPU or DPD sequences. These two sequences alternate in the anion topology, and are separated by R chains, giving the sequence RUPURDPURUPU...

Curite anion-topology

The structure of curite (Taylor et al. 1981) contains sheets based upon the curite anion-topology (Fig. 5e), which cannot be described as a simple chain-stacking sequence using only the U, D, P and R chains (Fig. 5d). A chain with pentagons, triangles and squares is required (Fig. 4b). It has a directional sense owing to the presence of an arrowhead (a pentagon and a triangle sharing an edge), and is designated Uᵐ and Dᵐ, for up and down (modified) pointing chains, respectively. The curite anion-topology can be characterized by the chain-stacking sequence UᵐDᵐU... (Fig. 5e).
The $\beta$–U$_3$O$_8$ sheet anion-topology (Fig. 5f) is the basis of the sheets in ianthinite (Burns et al. 1997a), as well as the sheets in $\beta$–U$_3$O$_8$. This topology can conveniently be described using a chain-stacking sequence involving U, D and R chains with the repeat sequence DRUDRU...

Wölsendorfite anion-topology

The $\beta$–U$_3$O$_8$ sheet anion-topology (Fig. 5f) is the basis of the sheets in ianthinite (Burns et al. 1997a), as well as the sheets in $\beta$–U$_3$O$_8$. This topology can conveniently be described using a chain-stacking sequence involving U, D and R chains with the repeat sequence DRUDRU...

Observations concerning chain-stacking sequences

Miller et al. (1996) used chain-stacking sequences to develop anion topologies that occur in uranyl oxide hydrates for which structures are known, and demonstrated that the approach could be used to predict new topologies. An interesting aspect of the chain-stacking sequences noted by Miller et al. (1996) is that P chains are always adjacent to either U or D chains in topologies, and the chains on either side of the P chain always have the same orientation, i.e., both are either U or D. The wölsendorfite and vandendriesscheite anion-topologies, discovered since the work of Miller et al. (1996), exhibit the same relationship between P, U and D chains (Burns 1997, 1999a). Miller et al. (1996) noted that the degree of structural misfit is identical within the UPU,
DPD and UPD sequences; thus it is unclear why the UPU and DPD sequences dominate.

New anion topology in KUOH

The anion topology of KUOH, derived using the methods of Burns et al. (1996), is presented in Figure 6. The U, D, and P chains, as well as a distorted R chain, are required to construct the anion topology, which has not been previously reported. The chain stacking-sequence is DUPDPUDRDUPDPUDR... Although this anion topology contains chains found in other topologies, the arrangement of the chains is unique. Segments of alternating UPDPU and DRD occur in the chain-stacking sequence. The occurrence of the UPDPU sequence is remarkable, as it is the first occurrence of P chains flanked by U or D chains of opposite directional senses. This finding confirms the observation of Miller et al. (1996) that P chains need not be flanked by U or D chains of the same orientation. In addition, the occurrence of an R chain flanked by two D (or U) chains is unique to this topology.

Related mineral species

There are three K uranyl oxide hydrates that have been described as minerals: compeignacite, K₂[(UO₂)₃O₂(OH)₃]₂(H₂O)₇ (Protas 1964, Burns 1998c), agrinierite, K₃(Ca,Sr)((UO₂)₂O₃(OH)₂)·2.5H₂O (Cesbron et al. 1972, Cahill & Burns 2000), and rameuite, K₂CaU₆O₂₇·9H₂O (Cesbron et al. 1972). Each of these has been reported from the Margnac deposit, Haute-Vienne, Massif Central, France, and formed following the alteration of uraninite in K-rich rocks. Elton et al. (1995) also reported the occurrence of compeignacite from Cornwall, England. On the basis of the chemical similarity of KUOH and the known K uranyl oxide hydrate minerals, nothing is apparent that precludes KUOH from occurring as a mineral in a similar geological environment.

Conclusion

The new topology found in KUOH again demonstrates the amazing complexity possible in sheets of uranyl polyhedra, and supports the hypothesis that many more sheets may exist. It is conceivable that many of these sheets are energetically similar, thus their occurrences may be sensitive indicators of geochemical conditions at the time of crystal growth.

Acknowledgements

This research was funded by the Environmental Management Sciences Program of the United States Department of Energy (DE–FG07–97ER14820).
manuscript was improved following reviews provided by two anonymous referees, and editorial work by R.F. Martin.

REFERENCES


Received May 26, 1999, revised manuscript accepted December 15, 1999.