STUDIES OF URANIUM MINERALS (X): URANOPILITE*

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ABSTRACT

The status of the many natural uranium sulfates that have been reported is reviewed. Only three appear to be valid species: uranopilite, zippeite and johannite. New optical, chemical and dehydration data are given for uranopilite together with *x*-ray powder data for uranopilite, zippeite and johannite. New localities for uranopilite are Great Bear Lake and Hottah Lake, N.W.T., Canada. Beta-uranopilite of Novacek (1935) probably is not a dehydration product of uranopilite and is a doubtful species.

INTRODUCTION

The descriptive mineralogy and nomenclature of the natural uranium sulfates presents many difficulties. Nineteen different names, tabulated below, have already been given as species or generic designations for uranium sulfates, but only three of these substances, *johannite, zippeite* and *uranopilite*, can be said to be adequately defined and recognizable on the basis of existing data.

Alpha-uranopilite
Basisches Schwefelsaures Uranoxyd
Basisches Uransulphat
Beta-uranopilite
Calciouraconite
Cuprozippeite
Dauberite
Johannite
Medjidite
Uraconise
Uraconite
Uranblüthe
Urangrün
Uranochalcite
Uranocker
Uranopilite
Uranvitriol
Voglianite
Zippeite

Further, descriptions have been given in the literature of unnamed sulfates whose properties do not correspond to any of these minerals so far as known. Johannite is a relatively well-established species, whose *x*-ray crystallography has been described by Hurlbut (1950) in a preceding paper of this series. The species rank of uranopilite and of zippeite was first put on adequate grounds by the careful study of Novacek (1935), and a further description of uranopilite is given in the present paper. *X*-ray powder data for zippeite, obtained from a study of both natural specimens and analyzed synthetic material, and for johannite also are given here. Some of the localities cited for zippeite in the literature are not well authenticated and may refer to uranopilite or some other uranium sulfate.

Very little is known of the other reported uranium sulfates. Many were set forth as distinct species on the basis of chemical analyses which were made by faulty analytical procedures, or which were made on

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mixtures. The status of *uranochalcite* and of *voglianite* has been discussed by Novacek. The original descriptions by Vogl (1857) of these substances and later notices by others of minerals ascribed thereto are inadequate to establish their species validity and a complete re-examination of the type materials is needed. Such material does not appear to be extant. Novacek, however, examined all of the specimens reputedly of uranochalcite and voglianite that were available in various museum collections in Czechoslovakia and Austria. He found that these specimens were not of uranium sulfate at all but for the most part were composed of the copper-uranium silicate cuprosklodowskite. One specimen was a uranium phosphate and another proved to be langite or herrengrundite. Of the material here available, two specimens supposedly of voglianite proved to be identical with liebigite, and five specimens supposedly of uranochalcite variously were metatorbernite, cuprosklodowskite and uranospinite.

The substances called *uraconise* (uraconite) by Beudant (1832) and *medjidite* by J. L. Smith (1848) are only names on paper. Beudant's uraconise is classed with the uranium sulfates by most modern authors. His description states only that the substance is a yellow powder and a specific locality is not given. The composition is given as a hydrated oxide of uranium, on the basis of a recalculated analysis affording oxygen 5.24 per cent, uranium 94.76, and an unknown amount of water. This scant information together with a statement that the mineral sometimes contains carbonic acid is repeated in the 3rd (1850) edition of Dana's *System of Mineralogy*, where the name is given as a synonym of zippeite, and in the 4th (1854) edition where it is given as a synonym of uranochre. In the 5th (1868) edition, Dana applied the name *uraconite* to an ill-defined uranium sulfate from Joachimsthal analyzed poorly by Lindacker (1857) and called uranocker by Vogl (1857). The 6th (1892) edition contains the same account. Uraconite has the same meaning as uraconise, and since Dana condemned Beudant's practice of ending mineral names in *ise*, it appears that uraconite was intended as a substitute for that name. Nevertheless, Dana cites Beudant's uraconise as a doubtful synonym of uraconite, and remarks that the composition of uraconise is unknown. In any case, the substance analyzed by Lindacker, and on which the name uraconite is based by Dana, is according to Novacek (1935) only zippeite. Uraconite is essentially a name without a mineral and should be abandoned. It is a common designation on museum labels; the specimens usually turn out to be zippeite or uranopilite.

Occurrences of uranium sulfates whose true identity is quite unknown have been mentioned under the name uraconite by Gordon (1922)

[Avondale, Chester and Leiperville in Delaware Co. and Fairmount Park in Philadelphia, all in Pennsylvania], Johnston (1915) [Madoc, Hastings Co., and Snowdon, Haliburton Co. in Ontario, Canada], Luquer (1904) [Bedford, Westchester Co., New York], James (1947) [Cornwall, England], and Sohon (1951) [Branchville and Middletown, Conn.] Larsen (1921) gives an optical description of an unanalyzed but apparently distinct sulfate from Gilpin County, Colorado, which he refers to as uraconite, and Koritnig (1939) refers to an undetermined sulfate from Schwag near Trahütten, Styria, Austria, as uraconite or zippeite. Rickard (1895) mentions yellow uranium ocher in an oxidized gold quartz vein containing uraninite [?] in the Rathgeb Mine, San Andreas, Calaveras Co., California, and some later authors have cited this occurrence as of uraconite.

Elsewhere in his book, Beudant mentions a *sulfate verte d'urane* (later to become known as johannite) and a yellow, earthy *sous-sulfate d'urane* whose true identity is not now known. *Medjidite* of Smith (1848) is an amber-brown massive material that was said on the basis of qualitative tests to be a hydrated sulfate of uranium and calcium, and other characterizing data are lacking. Two specimens of uncertain authenticity labelled medjidite that were examined in the course of this study proved to contain only uraninite and liebigite. The *uranocker* of Vogl (1857) from Joachimsthal, which has also been classed by some under the name uraconite, is an earthy or scaly, lemon-yellow substance which from the evidence of two discordant analyses appears to be identical with zippeite. *Dauberite* of Adam (1869) is a synonym of zippeite and *alpha-uranopilite* of Foshag (1935) is a synonym of beta-uranopilite. The names *uranvitriol*, *urangrün* and *uranblüthe* have been used more or less as generic terms for uranium sulfates in the German literature.

All of the above-mentioned names are devoid of any exact meaning and can hardly be applied to new occurrences of specific uranium sulfates in lack of a definitive re-description of the original materials. It seems desirable and justifiable to completely abandon these names, and to relieve any future investigator of the responsibility of resolving the earlier nomenclature before proposing a new species that differs from the three well-established species that are presently known.

Several additional uranium sulfates have been reported that may have a real existence. Novacek (1935) briefly described a natural fibrous sulfate close in composition to uranopilite under the name *beta-uranopilite* (meta-uranopilite). The status of this mineral is discussed beyond in connection with a dehydration study of uranopilite. Boldyrev (1935), in a publication not available to the writer, has used the names *cuprozippeite* and *calciouraconite* for minerals said to be near $Cu(UO_2)_3$

$(SO_4)_3(OH)_2.11H_2O$ and $Ca(UO_2)_4(SO_4)_2(OH)_6.20H_2O$, respectively. These names recall the old analyses of zippeite and uranopilite that contain CaO and CuO due to admixture.

URANOPILITE

The identification of the mineral here described as uranopilite is based principally on the definition of this species given by Novacek (1935), to whose excellent paper reference should be made both for a history of the name and for a summary of earlier recorded optical and chemical data.

Uranopilite is a secondary mineral and in part at least is of recent formation as an efflorescence on the walls of mine workings. It commonly occurs as a coating on oxidized vein material in which uraninite and sulfides were primary constituents. Gypsum and zippeite are common associates. The mineral forms fragile, small-botryoidal crusts or isolated, globular to reniform incrusting masses of small size. These are composed of tiny and sometimes microscopic lath- or needle-like crystals that form felted aggregates. The measured specific gravity is 3.96 (Wheal Owles, Cornwall). The hardness cannot be determined accurately because of the fibrous nature of the material but apparently is quite low. The color is bright lemon yellow, sometimes straw yellow in compact crusts, and the luster is faintly silky due to the fibrous character. Uranopilite shows the bright yellow-green fluorescence characteristic of uranyl compounds in both long- and short-wave ultraviolet light. In the hand specimen, the fine-fibrous and silky appearance of uranopilite contrasts with the fine-granular to earthy and rather dull appearance of zippeite. The color of the two minerals often is quite similar, but zippeite tends more toward deep yellow to orange-yellow tones. A distinction between uranopilite and zippeite is easily made by either optical or *x*-ray diffraction tests. Fluorescence is not a reliable means of discrimination between the two minerals. Uranopilite uniformly fluoresces a bright yellow-green, but zippeite varies from yellow-green (especially in fresh synthetic material obtained by adding very dilute NH_4OH to a dilute water solution of uranyl sulfate until a precipitate barely forms and then allowing to stand) through dull yellow green and dull yellowish olive green, and some material apparently fluoresces only weakly, if at all. Uranopilite is insoluble in cold water. It is easily soluble in dilute acids.

Under the microscope the mineral appears as elongated laths and needles with the optical orientation shown in Fig. 1. The laths sometimes are seen to be doubly terminated by oblique faces. The mineral appears to be monoclinic, with the elongation along [001] and the flattening on {010}. There is a good cleavage on {010}. Sections parallel {100} show parallel extinction with abnormal blue interference colors. The dispersion is rather strong, with $r>v$. The indices of refraction are tabulated below. They represent the range of numerous values here measured on air-dried, Museum specimens. Concordant optical data are given by Novacek (1935) and Buttgenbach (1935, 1947). The optical data given in the tables of Larsen and Berman (1934) for zippeite actually refer to uranopilite; the source of this error has here been traced back to a borrowed specimen that was erroneously labelled. It is shown beyond that the optical properties of uranopilite vary widely accompanying loss of water by heating or desiccation.

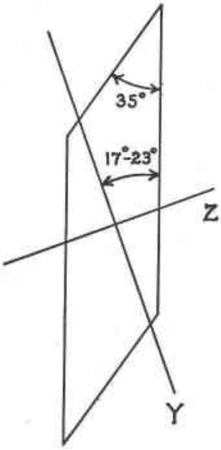

FIG. 1. Optical Orientation of Uranopilite.

Optical Properties of Uranopilite

Indices	Pleochroism	
$nX=1.621$ to 1.623	Colorless	Biaxial positive (+)
$nY=1.623$ to 1.625	Yellow	$r>v$, strong
$nZ=1.632$ to 1.634	Yellow	$Y\wedge c=17°$ to $23°$

The x-ray powder spacings of uranopilite, zippeite, and johannite are listed in Table 1.

Chemistry. Uranopilite is a hydrated basic uranyl sulfate. Novacek (1935) derived the ratios $6UO_3\cdot SO_3\cdot 16$ or $17H_2O$, corresponding to $(UO_2)_6(SO_4)(OH)_{10}\cdot 11$ or $12H_2O$, from the six new analyses reported by him. The three earlier analyses of Schulze (1882) and Dauber (1854) are of inferior quality. A later analysis by Buttgenbach (1935) of material from the Belgian Congo is close to $(UO_2)_5(SO_4)(OH)_8\cdot 10H_2O$, and a recent analysis of uranopilite from France reported by Branche, Chervet and Guillemin (1952) approximates to $(UO_2)_6(SO_4)(OH)_{10}$

TABLE 1. *X*-RAY POWDER SPACING DATA FOR URANOPILITE, ZIPPEITE AND JOHANNITE

Copper radiation, nickel filter, in Å($\lambda = 1.5418$)

Uranopilite		Zippeite		Johannite	
d	*I*	*d*	*I*	*d*	*I*
9.18	8	8.60	3	7.81	10
8.21	2	7.06	10	6.24	9
7.12	10	6.42	1	5.61	3
5.99	2	5.45	4	4.89	1
5.51	4	4.29	3	4.42	3
5.13	1	3.89	2	4.22	1
4.76	1	3.66	3	3.89	4
4.61	1	3.51	9	3.74	3
4.28	8	3.12	8	3.53	2
4.02	1	2.87	4	3.42	5
3.86	2	2.72	2	3.24	1
3.65	5	2.65	4	3.13	4
3.48	2	2.478	3	3.06	4
3.31	4	2.340	2	2.93	2
3.08	1	2.216	5	2.806	3
2.99	3	2.143	3	2.687	3
2.90	3	2.096	2	2.588	3
2.75	1	2.050	4	2.455	2
2.69	2	1.944	3	2.385	1
2.585	2	1.879	1	2.321	1
2.417	1	1.865	1	2.245	2
2.356	1	1.835	3	2.199	1
2.275	1	1.749	5	2.143	2
2.243	1	1.698	5	2.096	2
2.190	3	1.668	1	2.048	1
2.126	1	1.632	1	2.013	2
2.075	1	1.601	1	1.929	3
2.040	1	1.565	4	1.895	3
2.013	1	1.523	2	1.852	4
1.932	2	1.496	2	1.803	1
1.880	2	1.463	1	1.770	2
1.826	3	1.426	2	1.711	2
1.757	3	1.402	3	1.678	1
1.732	1	1.371	2	1.599	2
1.700	2	1.329	2	1.571	1
1.674	1			1.554	1
1.638	1			1.531	3
1.604	1			1.490	3
				1.463	2
				1.439	1

$\cdot 13H_2O$. A new analysis of uranopilite from the Wheal Owles, Cornwall, is cited below. This analysis, made on a 200 mg. sample, agrees well with earlier analyses in content of UO_3 but contains somewhat less SO_3 and has a slightly greater content of H_2O, being comparable to one of the analyses of Schulze (1882) in this regard. The ratios of the analysis are closer to $(UO_3)_7(SO_4)(OH)_{12}\cdot 15H_2O$ than to the formula of Novacek. The divergence among the reported analyses in the $UO_3\cdot SO_3$ ratio probably is due to analytical error. The water of crystallization is in part loosely held, as discussed beyond, and its variation probably reflects the temperature and humidity at which the sample was held prior to analysis. The CaO reported in some of the analyses is due to admixed gypsum. The formula advanced by Novacek and cited above probably best represents the composition of the mineral, although the ideal water content may be slightly higher.

URANOPILITE. WHEAL OWLES, ST. JUST, CORNWALL

UO_3	Fe_2O_3	Al_2O_3	CaO	SO_3	H_2O	Insol.	Total	G
79.44	0.05	nil	nil	3.30	15.68	1.72	100.19	3.96

Analyst: R. Meyerowitz, U. S. Geological Survey, 1949.

Localities. The following localities for uranopilite were established by Novacek on optical and analytical evidence: Johanngeorgenstadt, Saxony; Příbram, Bohemia; Joachimsthal, Saxony, St. Just, Cornwall. Specimens from all of these localities were examined in the present study and the identification was verified by optical and *x*-ray study. One very fine specimen from the Wheal Owles, St. Just, Cornwall, comprised a deeply altered mass of uraninite-bearing vein material the size and almost the color of an orange that was incrusted by uranopilite. The original handwritten label of this specimen reads: *Uraconite, Uran-ochre. From Wheal Owles, nr. St. Just, Cornwall. Evidently a secondary product on Pitchblende. Hydrous oxide of uranium with variable proportion of sulfuric acid. An indefinite mixture. This is a very fine specimen. Bought of Gregory, December 19, 1885. £1.10. J.P.*[*James Percy*] *December 20, 1885.* James (1947) mentions zippeite as occurring at the Wheal Owles among other places in Cornwall, and Greg and Lettsom (1858) mention several localities for zippeite (which they consider an oxide) in Cornwall; probably both zippeite and uranopilite were included by these authors under the single name. Uranopilite also has been identified by George (1949) in ore samples from the Urgeirica mine, Canas de Senhorim parish, Viseu district, Portugal. An optical description and analysis of the uranopilite from Shinkolobwe in the Belgian Congo has been given by Buttgenbach (1935–1947). Two occurrences in France, at La Crouzille, Haute-Vienne, and Grury, Saône-et-Loire (with analysis) are cited by Branche, Chervet and Guillemin (1952). Two new occurrences of uranopilite were found during the present study. At Great Bear Lake, Canada, it occurs very sparsely as thin, felt-like crusts and films, locally small botryoidal, on massive pitchblende. The mineral is relatively dense and fine-grained, with a straw-yellow to pale yellow color and a dull-silky luster. It is associated with minor amounts of zippeite as golden-yellow crusts, tiny radial-fibrous globules of johannite, fourmarierite, and erythrite. Uranopilite also has been identified as thin coatings on altered pitchblende from Hottah Lake, N.W.T., Canada. This material, like that from Great Bear Lake, is relatively fine-grained with a pale yellow color and a felt-like structure.

Dehydration. In the course of his study of uranopilite, Novacek (1935) observed that laths of this substance when kept embedded in Canada balsam for some time changed in color from yellow to grayish brown. The extinction also became parallel and the birefringence decreased almost to zero. These changes were ascribed to dehydration. Later, Novacek (1942) made a dehydration study of analyzed uranopilite from Joachimsthal. His data show that of the (total) $16H_2O$ present in this material, $6H_2O$ are lost sharply between 60°–70° and the remainder is lost gradually up to about 250°–300°. Over about 250°–270° the SO_3 begins to be lost as well. The divergent results of Buttgenbach (1935) were explained by Novacek as due to confusion in the temperature scales employed, Centigrade and Reamur.

Further observations were made here on the dehydration of the analyzed Wheal Owles material. When heated in air at 72°, 112° or 152°, water is lost rapidly. The loss at 72° was found to be 5.2 weight per cent, corresponding to the loss found by Novacek at 60–70°. When heated to bright redness the mineral decomposes completely leaving U_3O_8. The color of the mineral changes during dehydration from the original lemon-yellow to orange or, at higher temperatures, to deep orange and reddish orange. The mineral simultaneously loses its property of fluorescing in long- and short-wave ultraviolet radiation. There are marked accompanying optical changes. The extinction becomes parallel, the pleochroism is less marked and the indices of refraction increase to values depending on the temperature: $nY = nZ \sim 1.71$ at 72°, $nY = nZ \sim 1.84$ at 112°, $nY = nZ \sim 1.89$ at 152°. The birefringence also decreases from the initial value of 0.011 and some grains become almost isotropic at higher temperatures. There is a small but noticeable variation in both birefringence and indices between different grains in the heated samples, due to lack of attainment of equilibrium, and it is not possible to measure all of the indices on any single grain. The heated samples give weak and very diffuse *x*-ray powder patterns.

When the heated samples are held at room temperature in air or are immersed in cold water the original water content is very quickly restored. The original color and fluorescence are simultaneously regained, and the rehydrated material gives a sharp *x*-ray pattern of uranopilite. The original optical properties, including inclined extinction, also are restored.

Meta-uranopilite. The name beta-uranopilite* was given by Novacek (1935) to a needle-to lath-like mineral of a grayish, dirty green or brownish tint which he considered to have probably formed by the natural dehydration of uranopilite. An analysis in which Ca and Cu could not be determined for lack of sufficient material gave UO_3 82.40, Fe_2O_3 2.03, SO_3 4.17, H_2O 9.40; total 98.00. The iron oxide presumably is due to admixture of limonite. From this analysis was derived the tentative formula $6UO_3 \cdot SO_3 \cdot 10H_2O$ or $(UO_2)_6(SO_4)(OH)_{10} \cdot 5H_2O$. Only a single specimen from Joachimsthal, Bohemia, is known. The mineral is biaxial negative with parallel extinction with Y parallel to the elongation and X perpendicular to the flattening of the laths. The indices of refraction are $nX=1.72$, $nY=1.76$, $nZ=1.76$, and pleochroism is not perceptible.

No mineral corresponding to meta-uranopilite was found among the numerous specimens of fibrous uranium sulfates here examined. One of the studied specimens from Joachimsthal, however, shows a yellow, coarsely fibrous mineral, admixed with uranopilite, which has optical properties close to those reported for meta-uranopilite. This material is biaxial negative, with $nX=1.685$ (colorless), $nY=nZ=1.754$ to 1.764 (yellow). Extinction parallel with X perpendicular to the elongation. The *x*-ray pattern, however, is identical with that of becquerelite although the optical properties diverge considerably from those of crystallized becquerelite from the Belgian Congo. The material may be a pseudomorph after a fibrous sulfate. It may be noted in this connection that becquerelite, a mixture of becquerelite with zippeite, or zippeite alone is formed, depending on the pH, when uranyl sulfate solution is partly neutralized by NH_4OH and allowed to stand.†

The status of meta-uranopilite is uncertain, and further work on the mineral is desirable. It now appears very unlikely that it is a dehydration product of uranopilite as suggested by Novacek. Uranopilite, as shown above, can be dehydrated to a point where the indices of refraction are much higher than those of meta-uranopilite and the water content considerably less. This material, however, differs from meta-uranopilite in

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* The name meta-uranopilite is preferable, since the prefix beta is generally applied to designate instances of polymorphism, as in uranophane and beta-uranophane, and meta to cases of alteration or dehydration, as in torbernite and meta-torbernite.

† Private communication from Prof. J. W. Gruner, University of Minnesota, 1950.

having a much lower birefringence and in rehydrating immediately to normal uranopilite when exposed to air at room temperature.

REFERENCES

ADAM, G. J., Tableau Minéralogique, Paris, 64 (1869).

BEUDANT, F. S., Traité élémentaire de Minéralogie, 2nd ed., Paris, **2,** 672 (1832).

BOLDYREV, A. K., Course of Descriptive Mineralogy, Leningrad and Moscow, **3,** 83 (1935) [cited by Hey, M. H.: Chemical Index of Minerals, London, 256, 257 (1950)].

BRANCHE, G., CHERVET, J., AND GUILLEMIN, C., *Bull. Soc. Min.*, **74,** 457 (1952).

BUTTGENBACH, H., Les Minéraux de Belgique et du Congo Belge, Liège, 515 (1947); *Mém. Inst. roy. colon. belge*, **6,** 449 (1935).

DAUBER, H., *Ann. Phys. Chem.*, ser. 4, **92,** 237 (1854).

FOSHAG, W. F., *Am. Mineral.*, **20,** 813 (1935).

GEORGE, D'A., *U. S. Atomic Energy Comm. Rept.* **RMO-563,173** (1949).

GORDON, S. G., Mineralogy of Pennsylvania: *Acad. Nat. Sci. Philadelphia, Spec. Publ.* no. **1,** 148 (1922).

GREG, R. P., AND LETTSOM, W. G., Manual of the Mineralogy of Great Britain and Ireland, London, 382 (1858).

HURLBUT, C. S., *Am. Mineral.*, **35,** 531 (1950).

JAMES, C. C., *Trans. Royal Geol. Soc. Cornwall*, **17,** Pt. 5, 256 (1947).

JOHNSTON, R. A. A., *Canada Dept. Mines, Geol. Survey, Mem.*, **74,** 230 (1915).

KORITNIG, S., *Zbl. Min.*, 116 (1939A).

LARSEN, E. S., *U. S. Geol. Surv., Bull.*, **679,** 159 (1921).

LARSEN, E. S., AND BERMAN, H., *U. S. Geol. Surv., Bull.* **848,** 112, 113 (1934).

LUQUER, L. M., *Am. Geol.*, **33,** 17 (1904).

NOVACEK, R., *České Spol. Nauk, Třida Mat.-Přiro., Věstník* no. **7,** (1935); *ibid.*, no. **17** (1941).

RICKARD, T. A., *Colorado Sci. Soc. Proc.*, **4,** 329 (1895).

SCHULZE analysis cited in Weisbach, A., *Jb. Min.*, **II,** 249 (1882).

SMITH, J. L., *Am. J. Sci.*, **5,** 337 (1848).

SOHON, J. A., Connecticut Minerals, Their Properties and Occurrence: *Conn. Geol. and Nat. Hist. Survey, Bull.* **77,** (1951).

VOGL, J. F., Gangverhältnisse und Mineralreichthum Joachimsthals, Teplitz (1857).

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