

THE AMERICAN MINERALOGIST

JOURNAL OF THE MINERALOGICAL SOCIETY OF AMERICA

Vol. 41

JULY-AUGUST, 1956

Nos. 7 and 8

MINERAL COMPOSITION OF GUMMITE*†

CLIFFORD FRONDEL, *Harvard University, Cambridge, Mass.,
and U. S. Geological Survey, Washington, D.C.*

ABSTRACT

The name gummite has been widely used for more than 100 years as a generic term to designate fine-grained yellow to orange-red alteration products of uraninite whose true identity is unknown. A study of about 100 specimens of gummite from world-wide localities has been made by x -ray, optical, and chemical methods. It proved possible to identify almost all of the specimens with already known uranium minerals.

Gummite typically occurs as an alteration product of uraninite crystals in pegmatite. Such specimens show a characteristic sequence of alteration products: (1) A central core of black or brownish-black uraninite. (2) A surrounding zone, yellow to orange-red, composed chiefly of hydrated lead uranyl oxides. This zone constitutes the traditional gummite. It is principally composed of fourmarierite, vandendriesscheite and two unidentified phases (Mineral *A* and Mineral *C*). Less common constituents are clarkeite, becquerelite, curite, and schoepite. (3) An outer silicate zone. This usually is dense with a greenish-yellow color and is composed of uranophane or beta-uranophane; it is sometimes soft and earthy with a straw-yellow to pale-brown color and is then usually composed of kasolite or an unidentified phase (Mineral *B*). Sodydyite and sklodowskite occur rarely.

There are minor variations in the above general sequence. In some specimens the core may be orange-red gummite without residual uraninite or the original uraninite crystal may be wholly converted to silicates. Other specimens show clarkeite as the core, or an intermediate zone of clarkeite may form between the central uraninite and the orange-red zone of hydrated oxides. The uranyl carbonate rutherfordine has been observed as the outermost zone in some specimens, usually more or less admixed with uranyl silicates.

Chemically the formation of the minerals of the orange-red gummite zone at the expense of the original uraninite is characterized by oxidation of U^4 to U^6 and hydration. There is usually little or no addition of material other than H_2O at this stage, but leaching of U relative to Pb may be a factor. The Pb in the minerals of this zone is the original radiogenic lead of the uraninite. The outermost silicate zone is due to reaction of the hydrated oxides of the gummite zone with meteoric or possibly hydrothermal waters carrying silica and usually also calcium. The Pb may be removed at this stage, together with uranium in part, with the formation of uranophane, beta-uranophane, or soddyite, or it may be retained in part or entirely with the formation of Mineral *B* or kasolite. The Th and rare earths contained in the original uraninite usually are wholly removed during the formation of the silicates.

* Contribution from the Department of Mineralogy and Petrography, Harvard University, No. 361; Studies on Uranium Minerals, XI.

† Publication authorized by the Director, U. S. Geological Survey.

There is also a further stage of alteration of the uraninite of pegmatites, in which the substance of the pseudomorphs is leached away and is either dispersed or redeposited nearby in cracks in the matrix. The uranium minerals that occur in this way are chiefly the phosphates meta-autunite, parsonsite, and phosphuranylite, and the silicates uranophane, beta-uranophane, kasolite, and Mineral *B*. The alteration of uraninite in vein or other non-pegmatitic deposits follows the same general sequence of alteration described, both on a hand specimen and geologic scale. In the presence of abundant sulfides, however, a different assemblage of secondary minerals is formed. This is characterized by the formation of zippeite, uranopilite, johannite, and gypsum, and there may be extensive leaching of uranium in the ground waters.

HISTORY AND NOMENCLATURE OF GUMMITE

Gummite is a vague term that has been and is used in various ways. Probably its commonest present-day use is as a generic term for gumlike or dense fine-grained pseudomorphic alteration products of uraninite whose true identity is not known. The term ordinarily is employed in lack of any knowledge of the composition other than that uranium is a major constituent. It also has been applied, as in the 7th edition of Dana's System of Mineralogy, in a more restricted chemical sense to substances that are essentially hydrated oxides of uranium, whose true identity is unknown, that represent the final stages of oxidation and hydration of uraninite. George (1949) states that gummite is a generic term applied to the usually orange or orange-yellow alteration products of uraninite or pitchblende. Gummite is regarded by Hey (1950) as an indefinite generic term analogous to limonite and wad.

Gummite is also loosely used without regard either to the pseudomorphic nature of the material or to its chemical composition, provided usually that the material is orange-red to yellowish in color and dense in appearance, and the matter thus grades into the problem of unidentified uranium minerals in general. The color of the material commonly is orange-red, orange or yellow, but may be greenish yellow, brown, or other colors depending on the identity and, in part, the state of subdivision of the mineral or minerals present. A gumlike or resinous appearance was emphasized in the early descriptions and synonymy of this substance, but this feature has not been recognized as a necessary criterion.

The name gummite has not been used always in a generic sense. The first analyzed and clearly described gummite—the *hyacinthrothes Pechuran* of Freiesleben (1817) and Kersten (1832) from Johanngeorgenstadt, so-called in distinction from *schwarzes Pechuran*, or pitchblende, of which it was said to be an alteration—was said to be opaline and resembling amber in appearance. Kersten considered the substance to be a distinct species, and gave the formula as $\text{Ca}_3(\text{PO}_4)_2 \cdot 4\text{UO}_3 \cdot 9\text{H}_2\text{O}$. Later, a number of more or less similar ill-defined substances were described under various

given names in the belief that they were distinct species. Some of these substances appear to be essentially oxidized uraninite, others are hydrated oxidation products thereof, and many appear to have been mixtures with uranophane and other secondary uranium minerals. A few of these names have found their way into the synonymy of uraninite and others are included in the complex group of unidentified substances now called gummite. The name gummite itself was first introduced by J. D. Dana in 1868 as a species designation for an impure hydrated uranium oxide. He included in it the material of Kersten together with the Uran-gummi and Uranisches Gummi-Erz of Breithaupt (1830, 1847) and the Phosphor-Gummit of Hermann (1859). The pittinite of Hermann (1859) and eliasite of Haidinger (1852) were classed as a hydrated uranium oxide species (eliasite) separate from both uraninite and gummite, but it was remarked by Dana that the species may not be distinct from gummite. In 1883, Von Foullon made a detailed microscopic and chemical examination of gummite in the sense of Dana, including therein pittinite, eliasite, and the coracite of Whitney (1849) and Genth (1857), and sought to show that the substance was a definite species with the composition $(\text{Pb}, \text{Ca}, \text{Ba})\text{U}_3\text{SiO}_{12} \cdot 6\text{H}_2\text{O}$. Strunz (1949) accepts this view in part. He considers that gummite is principally a gel-like, pseudomorphous alteration product of uraninite with the formula given by Von Foullon, but certainly also in part a mixture of different cryptocrystalline uranium minerals. This particular matter is discussed further below—where it is shown from a re-examination of Von Foullon's original specimens that his formula represents a mechanical mixture of a lead uranyl oxide with a calcium uranyl silicate.

E. S. Dana in the 1892 edition of the *System of Mineralogy* placed gummite as a numbered species in the Uranate Group and stated that it is an alteration product of uraninite of doubtful composition, with the implication that further study would characterize it as a distinct species. It is difficult to see how this could be effected. Evidence is lacking which permits gummite in the sense of Dana either to be identified with or distinguished from any of the many known uranium minerals within the range of the chemical analyses and descriptions of gummite that have been reported, nor are type specimens available whose re-study would arbitrarily resolve the problem, as the name is not based on the description of a particular specimen but was applied to a pre-existing group of minerals thought but never shown to be identical. The present tendency to use gummite as a generic term is a recognition of these difficulties.

Gummite together with limonite and wad are modern examples of the great pocket terms that contained much of ancient mineralogy. Among the latter may be mentioned bole, misy, and especially schorl, which once

included even the rudely hexagonal columns of basaltic lava. Schorl and the others were largely resolved into their component species in the decades immediately preceding and following 1800, with the advance of chemistry and morphological crystallography in that period. In the present day, gummite and its analogues virtually disappear as units of description if *x*-ray diffraction, optical, and thermal methods are applied. The difficulties obtaining with gummite in the past were largely due to the very fine-grained and often admixed nature of the material. It is of interest to note that only one mineral, uranophane, of the dozen or so that have now been identified as constituents of gummite pseudomorphs was known at the time of the 1892 edition of Dana's *System of Mineralogy*.

In the present study, a collection of about 100 specimens of gummite from world-wide localities was examined by *x*-ray, optical, and chemical methods. All of the specimens were partial or complete pseudomorphs after uraninite, and had the color, dense appearance, and other characters attributed to gummite. It proved possible to identify almost all of the specimens with already known uranium minerals. Brief descriptions of these minerals are given in a later section of this paper. A few unidentified minerals, apparently new species, also were found. Three of these, Minerals *A*, *B*, and *C*, are briefly described in the present report.

The optical properties of the minerals that occur in gummite or that are associated therewith are summarized in Table 3, appended. The *x*-ray power spacing data for these minerals are given in Table 4, appended.

CHEMICAL ANALYSES OF GUMMITE

Nonexistence of Gummite as a Species

The reported chemical analyses of gummite are listed in Table 1. A brief discussion may be given here of these analyses in advance of a detailed description of the mineralogy of the substance. Von Foullon (1883) cited all of the older analyses given in Table 1. He accepted the SiO_2 and CaO , both present in fairly constant amounts, as essential constituents and derived the formula $(\text{Pb}, \text{Ca}, \text{Ba})\text{U}_3\text{SiO}_{12} \cdot 6\text{H}_2\text{O}$ for gummite.

The silica reported in the analyses of gummite, however, appears, from the observations of Genth (1879) and much other evidence, to be due to admixture principally of uranophane or beta-uranophane, $\text{CaO} \cdot 2\text{SiO}_2 \cdot 2\text{UO}_3 \cdot 6\text{H}_2\text{O}$, and Von Foullon's interpretation of gummite as a definite species with the formula cited never found general acceptance. The writer has examined all of Von Foullon's specimens through the courtesy of Dr. A. Schiener, Curator of Mineralogy of the Natural History Museum, Vienna. The North Carolina specimens had been originally obtained by Von Foullon from W. E. Hidden [1853-1918], an American mineralogist. Unfortunately, it is not possible to correlate particular specimens with

TABLE 1. CHEMICAL ANALYSES OF GUMMITE.

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
CaO	3.19	3.04	2.05	0.90	4.54	3.87	3.09	2.26	1.37	0.48	6.00	2.1	10.36		1.64	tr.	1.10	4.60	3.80
MgO	1.04	0.92	1.08	2.16	0.85	tr.	2.20	0.55	0.12	0.27									
BaO	5.51	4.69	5.57	5.28	5.04	3.92	4.62	2.51	14.93	20.15			1.66						
PbO	0.41	1.06		0.29	8.64	7.25	6.63	4.54				4.0	6.39	0.71	6.93	5.20	10.92	7.90	6.67
Fe ₂ O ₃			0.53	0.86			1.17		0.14										
Al ₂ O ₃	74.79	74.50	75.20	77.99	63.38	66.57	61.33	68.45	60.36	73.20	72.00	77.4	68.20	81.32	82.19	80.97	78.13	62.91	60.88
UO ₃			0.12				0.84				2.30				tr.	tr.	0.15	1.34	
P ₂ O ₅	5.03	5.04	4.63	1.97	4.92	4.96	5.13	5.00	3.81	0.61	4.26	1.4	2.80	8.20	3.80	3.04	2.50	13.05	15.75
SiO ₂	9.86	9.94	10.54	8.90	10.24	11.86	10.68	10.06	9.42	5.33	14.73	9.3	9.45	7.29	6.10	8.80	7.00	11.00	11.00
H ₂ O				2.51	1.92	0.74	3.61	5.87	9.41		0.05	6.0	0.15	1.67	0.40	2.96	0.05	0.87	0.25
Total	99.83	99.19	99.72	100.86	99.53	99.17	99.30	99.24	99.56	100.04	99.36	100.2	99.01	99.19	101.06	100.97	99.70	100.43	99.19

- Orange red material. Mitchell County, North Carolina. Von Foulton (1883). Average of two analyses.
- Orange red material. Mitchell County, North Carolina. Von Foulton (1883).
- Orange red material. Flat Rock mine, Mitchell County, North Carolina. Genth (1879).
- Orange red material. Wiseman's mine, Spruce Pine, Mitchell County, North Carolina. Rees, Henderson, and Ponsjak (1931). Rem. is K₂O 0.86, Na₂O 0.51, ThO₃ and rare earths 1.06, insol. 0.08. Mineral C.
- "Elasticite," Joachimsthal, Bohemia. Von Foulton (1883) cites two additional very similar analyses. Rem. is Mn₂O₃.
- "Elasticite," Joachimsthal, Bohemia. Ragsky (1853). Rem. is FeO 1.09, CO₂ 2.52.
- "Pittinite," Joachimsthal, Bohemia. Hermann (1859). Rem. is Bi₂O₃ 2.67, insol. 3.20.
- Flame-scarlet material. Villeneuve, Quebec. Ellsworth (1930). Rem. is ThO₃ 7.66, Ce₂O₃ 0.14, Y₂O₃ 1.61, CO₂ undet.
- Brownish-yellow material. Kambove, Katanga. Schoep and de Leenherr (1937).
- Hyacinth-red material. Johannegeorgenstadt, Saxony. Kersten (1832). Rem. is Mn₂O₃.
- Brownish-red material. Mitchell County, North Carolina. Analysis by R. Meyrowitz, U. S. Geological Survey, 1954. Rem. is Na₂O 1.1, UO₃ 0.4, R₂O₃ (mostly Y and Al) 4.5, H₂O is H₂O+5.6, H₂O-3.7. Sp. Gr. 5.39. Mineral C.
- Orange-red material. Luiwishi, Katanga. Gillet analysis in Buttgenbach (1922). Sp. Gr. 5.08.
- Gummite. Alto do Tibiri, Paraitiba, Brazil. Rem. is UO₃. Florencio (1951).
- Orange material. La Chiquita mine, Sierra de Comenchingones, Córdoba, Argentina. Rem. is ThO₃ and rare earths. H₂O is loss on ignition. Chaudet analysis in Ahlfeld and Angeli (1948).
- Yellowish-orange material. Angel mine, Calamuchita, Córdoba, Argentina. Rem. is ThO₃ and rare earths 2.15, UO₃ 0.81, H₂O is loss on ignition. Sp. Gr. 4.10. Chaudet analysis in Ahlfeld and Angeli (1948).
- Yellow material. Tigre mine, Sierra de Comenchingones, Córdoba, Argentina. Rem. is ThO₃ and rare earths 0.05, H₂O is loss on ignition. Sp. Gr. 4.20. Chaudet analysis in Ahlfeld and Angeli (1948).
- Light-brown material. Tigre mine, Sierra de Comenchingones, Córdoba, Argentina. Rem. is ThO₃ and rare earths. H₂O is loss on ignition. Chaudet analysis in Ahlfeld and Angeli (1948).
- Yellow material. Tigre mine, Sierra de Comenchingones, Córdoba, Argentina. Rem. is ThO₃ and rare earths. H₂O is loss on ignition. Chaudet analysis in Ahlfeld and Angeli (1948).

the analyses cited by Von Foullon. X-ray and optical studies of his specimens from Mitchell County, North Carolina, including the materials of analyses 1 and 2 (Table 1), although these could not be specifically identified, show that they are variable mixtures of uranophane with vandendriesscheite or an unidentified lead uranyl oxide. Specimens of identical appearance from Mitchell County contained in other collections also were found to be mixtures of uranophane or beta-uranophane, with vandendriesscheite and, in some specimens, clarkeite, fourmarierite, and unidentified lead uranyl oxides. A sample of the gummite from Mitchell County analyzed by Ross, Henderson, and Posnjak (1931), cited in column 4 of Table 1, and supplied for examination by Dr. George Switzer of the U. S. National Museum, was found on x-ray and optical study to be an unidentified phase, Mineral C. This mineral is closely associated with uranophane, and the analyzed sample is described as of questionable homogeneity. A new analysis of a brownish-red gummite from Mitchell County is cited in column 12 of Table 1. This material also gave the x-ray pattern of Mineral C.

If the analyses of the material from Mitchell County cited in columns 1, 2, and 3 of Table 1 are recalculated after deduction of the SiO_2 as uranophane, $\text{CaO} \cdot 2\text{UO}_3 \cdot 2\text{SiO}_2 \cdot 6\text{H}_2\text{O}$, the remaining substance is seen from the data of Table 2 to have virtually the composition of vandendriesscheite. Analyses 1 and 2 have a slight excess of CaO over the requirements of the SiO_2 for uranophane, and analysis 3 a slight deficiency (3.19, 3.04, 2.05 present; 2.35, 2.35, 2.16 required). In the analyses as recalculated, all of the CaO and SiO_2 was deducted together with the amounts of UO_3 and H_2O required by the SiO_2 present. The Ba present in these analyses undoubtedly substitutes for Pb.

Analysis 11 by Kersten (1832) of the "hyacinthrothes Pechuran" from Johanngeorgenstadt is rather similar to those just discussed if it is assumed, as was suggested by Von Foullon (1883), that much of the Ca reported actually is Pb. Analyses 5, 6, and 7 of eliasite and pittinite from Joachimsthal apparently represent mixtures of limonite and uranophane with a lead uranyl oxide. Type specimens of eliasite and pittinite are not available for examination. However, one specimen from Joachimsthal of uraninite altered to orange-colored gummite was found here to be composed of fourmarierite and uranophane and another was composed of vandendriesscheite. Analysis 10 is close to curite in composition. Analysis 9 was made on admittedly inhomogenous material containing perhaps 25 per cent of another mineral that appears from its description to be uranophane. The deep color and high lead content of this material suggests that the gummite component was perhaps curite. Rigal (1938) cites a partial analysis of a very impure orange gummite from Argentina

that apparently contains about 5 per cent PbO and 80 per cent UO_3 . Chaudhuri (1944) found radiometrically that an orange-yellow gummite from Rajputana contained about 71 per cent UO_3 and 0.8 per cent ThO_2 . Krishnan (1948) cites partial analyses of impure gummite from Rajputana. Florencio (1951) has reported an analysis of a gummite pseudomorph after uraninite from Alto do Tibiri, Paraiba, Brazil, that apparently consists of a hydrated uranyl silicate, possible soddyite. The analysis is reported in column 14, Table 1. Five analyses of gummite and uranium ochre from Córdoba province, Argentina, reported by Ahlfeld and Angelelli (1948) are cited in columns 15 to 19, Table 1. The material

TABLE 2. RECALCULATED ANALYSIS OF GUMMITE

	1	2	3	4	5	6
BaO	1.7	1.5	1.6			
PbO	8.8	7.7	8.3	9.14	15.31	22.10
UO_3	81.0	82.0	80.5	82.00	78.51	75.52
H_2O	8.5	8.8	9.6	8.86	6.18	2.38
Total	100.0	100.0	100.0	100.00	100.00	100.00

1. Mitchell County, North Carolina. Von Foullon (1883). Analysis 1, Table 1, recalculated after deducting uranophane and Fe_2O_3 .
2. Mitchell County, North Carolina. Von Foullon (1883). Analysis 2, Table 1, recalculated after deducting uranophane and Fe_2O_3 .
3. Mitchell County, North Carolina. Genth (1879). Analysis 3, Table 1, recalculated after deducting uranophane and Al_2O_3 , P_2O_5 .
4. Vandendriesscheite. Theoretical composition, $\text{PbO} \cdot 7\text{UO}_3 \cdot 12\text{H}_2\text{O}$.
5. Fourmarierite. Theoretical composition, $\text{PbO} \cdot 4\text{UO}_3 \cdot 7\text{H}_2\text{O}$.
6. Curite. Theoretical composition, $3\text{PbO} \cdot 8\text{UO}_3 \cdot 4\text{H}_2\text{O}$.

occurs as alteration products of uraninite in pegmatites. The analyses resemble those of gummite in general, and apparently are mixtures with uranophane and, in analysis 19, possibly also quartz or other insoluble material. The presence of uranophane (?) is noted in material from Angel mine (analysis 16, which lacks CaO), and this mineral was identified in specimens of gummite from Córdoba examined by the writer.

The *x*-ray and chemical evidence indicates that the gummite from Mitchell County, North Carolina, is not a definite compound containing essential Ca and Si, as was believed by Von Foullon, but is a mixture of a calcium uranyl silicate, uranophane, with a lead uranyl oxide, usually vandendriesscheite. This view is strongly supported by the fact that similar specimens of gummite, from localities over the world, are found to be composed of lead uranyl oxides, usually vandendriesscheite,

Mineral *A*, Mineral *C*, or fourmarierite, admixed or associated with the silicates uranophane, beta-uranophane, kasolite, or Mineral *B*.

SEQUENTIAL ALTERATION OF URANINITE
Mineralogy

Gummite pseudomorphs after uraninite crystals show a very characteristic sequence of alteration products that has long been recognized. Brief descriptions have been given by Kerr (1877), Genth (1879), Hidden (1881), Von Foullon (1883), Hoffman (1901), Tipper (1919), Schoep (1921), Buttgenbach (1922), Hacquaert (1927), Ellsworth (1930), Ross, Henderson, and Posnjak (1931), Yagoda (1946), Sarkar and Sen (1946), Krishnan (1948), Bakken, Gleditsch, and Pappas (1948), and Page (1950). The best examples are afforded by the uraninite crystals found in pegmatites, but the same principles apply to uraninite in all of its occurrences. When a typical, partly altered crystal is broken across, one finds: (1) A veined or embayed central core of uraninite. The color may be black or, in relatively highly oxidized material, greenish black, brownish black, or dark brown. (2) A surrounding zone of orange-red, orange, brownish-red, brown, or yellow material that constitutes the traditional gummite. This material is dense in structure, although microscopically crystalline, and ranges from vitreous or gumlike to dull and earthy in appearance. In composition it consists chiefly of hydrated uranyl oxides, often containing considerable amounts of essential lead together with minor amounts of calcium and barium. The commonest mineral constituents are vandendriesscheite, fourmarierite, Mineral *A*, and Mineral *C*. Curite, becquerelite, and schoepite also have been identified, and several unidentified minerals have been found. (3) An outer zone of uranyl silicates. This zone generally is hard, dense, and microcrystalline with a greenish-yellow color and vitreous luster but may be soft and earthy with a straw-yellow color and dull luster. The hard, vitreous material often faithfully preserves the minute surface details of the original uraninite crystals. The silicate zone grades into the hydrated oxide zone and has been derived from it. The principal minerals of this zone are uranophane, beta-uranophane, and kasolite. Mineral *B*, soddyite, sklodowskite, and several unidentified minerals also occur.

This sequence may vary considerably in detail. Thus, the original uraninite may be completely oxidized leaving a pseudomorph of orange gummite, with a more or less thick rind of uranium silicate, or the pseudomorph may consist largely or entirely of uranium silicate. Further, additional more or less well marked zones may be present. Specimens from a few localities were found to have an irregular outer zone of the uranyl carbonate rutherfordine which graded inwardly into orange-red

gummite. Complete pseudomorphs of rutherfordine also were observed. Other specimens showed brown clarkeite as the core, or an intermediate zone of clarkeite was present between the uraninite core and the outer gummite and silicate zones. The gummite zone itself may be irregularly mottled or veined in orange-red, red, or yellow colors due to admixture of various minerals.

There is also a further stage of alteration of the uraninite of pegmatites, in which the substance of the pseudomorphs is leached away and is dispersed or redeposited in cracks in the matrix in the immediate neighborhood of the pseudomorphs. The secondary uranium minerals that occur in this way are chiefly the phosphates meta-autunite, parsonsite, and phosphuranylite and the silicates uranophane, beta-uranophane, kasolite, and Mineral *B*.

The alteration sequence just described seems to take place at essentially atmospheric conditions through the agency of meteoric waters. It also has been considered, apparently first by Ross, Henderson, and Posnjak (1931), that the alteration of the uraninite of pegmatites to gummite and silicates took place during the late hydrothermal stage of pegmatite formation. It may be noted in this connection, however, that the uraninite of various nonpegmatitic deposits, including the Colorado Plateau region, Katanga, Great Bear Lake, and Joachimsthal, undergoes an identical sequence of alterations under what are clearly weathering conditions. George (1949) also points out that in the uraninite vein deposits of Portugal, uranophane and beta-uranophane are much more abundant toward the surface and are there associated with typical supergene uranium minerals such as autunite and torbernite. According to Websky (1853, 1859), uranophane at its type locality, Kupferberg, Silesia, is formed by atmospheric influences on primary uraninite in the copper veins. Hayden (1914) remarks that uranium ocher (gummite) is common at the surface of pegmatites in the Gaya district, India, and passes into pure uraninite in depth (where a nodule 36 pounds in weight was found). This also is true in the New England pegmatites. The occurrence of uraninite in pegmatites has been discussed by Page (1950) and Quensel (1940). Lang (1952) mentions 27 occurrences of gummite in Canada, mostly in pegmatites.

The sequence of alteration of uraninite on a single-crystal scale is broadly paralleled by the alteration of vein deposits of uraninite on a geologic scale. Thus, in the Katanga deposits the uraninite alters first to hydrated uranyl oxides or lead uranyl oxides, principally becquerelite, schoepite, vandendriesscheite, and fourmarierite. These minerals occur chiefly as crystalline crusts and masses, involving deposition from solution, and are not pseudomorphous alteration products. Pseudomorphs

also occur but are rare. The very rare secondary mineral ianthinite, a hydrated oxide of quadrivalent uranium, is formed earlier than these hexivalent oxides and occurs as crystals in veinlets penetrating uraninite. The formation of the hydrated uranyl oxides is accompanied and followed by the deposition of hydrated uranyl phosphates and silicates, generally containing copper, calcium, magnesium, or lead in addition to uranium. These have formed as crusts and masses chiefly by the action of meteoric waters carrying Cu, Ca, Mg, phosphate, and silica on the earlier formed hydrated oxides. The phosphate apparently has been derived by meteoric waters from the country rock. Unfortunately there is no detailed account of the paragenesis of the secondary uranium minerals at Katanga in the available literature. Brief accounts are given by Robert (1940), Katz and Rabinowitch (1951), Everhart (1951), Thoreau and Terdonck (1933, 1935), Bain (1950), and Kohl (1954), and scattered remarks are found in the literature descriptive of individual species. The above sequence, recognized in the literature, is confirmed by the examination of Museum specimens. Uraninite deposits rich in sulfides generally alter directly to the sulfates zippeite, uranopilite, and johannite, and probably much of the uranium is lost in the meteoric circulation if the water is acid. The uraninite deposits of the Canadian shield, as a Great Bear Lake and Lake Athabaska, show only a very slight supergene alteration, chiefly to hydrated uranyl oxides and silicates.

The uraninite of the sandstone-type deposits of the Colorado Plateau generally is intimately associated with montroseite and other primary low-valence vanadium minerals and on oxidation passes to carnotite, tyuyamunite or other secondary uranyl vanadates. In a few instances, however, as at the Monument No. 2 mine, Monument Valley, Arizona, and the Delta mine, San Rafael Swell, Utah, uraninite is found superficially altered to becquerelite and schoepite; the lead-bearing oxides fourmarierite and vandendriesscheite are lacking except in traces. Lead-bearing secondary uranium minerals, including kasolite, curite, parsonsite, dewindtite, renardite and dumontite, are lacking in general in the Plateau deposits, in contrast to their abundant occurrence in the Belgian Congo. This is owing to the general absence of lead-containing minerals such as galena in the unoxidized ores and to the low content of radiogenic lead in the uraninite of the Plateau deposits.

Chemistry

The chemistry of the sequential alteration of uraninite crystals offers a number of interesting problems. In the initial stage of alteration, the uraninite itself oxidizes over a range of composition without destruction of its crystal structure. In this, it is well established that valence com-

pensation for the serial oxidation of U^4 to U^6 is effected by a coupled entrance of oxygen into the vacant eight-coordinated positions of the structure. The extent of oxidation that is possible before the structure breaks down is not known. It appears to be in the neighborhood of $UO_{2.3}$ (see Katz and Rabinowitch, 1951) or $UO_{2.6}$ (see Brooker and Nuffield, 1952). The effect that the presence of Pb, Th, and rare earths in substitution for U has on the extent of the oxidation is not known. A similar mechanism of compositional variation is known in other substances isostructural with UO_2 , including the substitution of Y, Th, or U^4 for Ca in CaF_2 ; La for Sr in SrF_2 ; and Bi for Pb in PbF_2 . In these instances F enters the interstitial positions to effect valence compensation. It also has been suggested that the content of U^6 in natural uraninite is due to an auto-oxidation involving radioactive decay processes independent of the environment (Ellsworth, 1925, and Khlopin, 1938). Attempts based on this assumption to use the U^4/U^6 ratio for age calculations have been unsatisfactory. It seems likely that the oxidation is a forerunner of the hydration and chemical reaction with traversing, oxidizing solutions that ultimately convert the uraninite completely into the minerals of the gummite and silicate zones. It is of interest to note that neither U_3O_8 nor the tetragonal polymorph of partly oxidized uraninite ($\sim UO_{2.4}$) have been found in nature, although both are stable, well-known compounds easily formed by the oxidation of UO_2 .

In the second stage of alteration, the uraninite is destroyed and its constituents are reorganized into the hydrated lead uranyl oxide minerals of the orange-red gummite zone. The change involves complete oxidation of the uranium to U^6 and hydration, without significant gain of material other than water, but possibly with leaching of uranium in part relative to lead. It is apparent from the examination of many specimens that the parent uraninite does not have to be oxidized to its limiting content of U^6 in the UO_2 structure-type before it converts to these new phases, although it may alter more readily if it is relatively highly oxidized. The radiogenic lead of the original uraninite seems to be wholly retained in the new minerals formed at this stage. The particular mineral or minerals formed here, whether schoepite or becquerelite (little or no lead), vanderiesscheite (9.1 per cent lead), fourmarierite (15.3 per cent lead), or curite (22.1 per cent lead) presumably reflects the amount of radiogenic lead available in the original uraninite. Leaching of uranium relative to lead is an added factor. No zoning of the minerals of this stage in point of lead content has been observed. The amount of radiogenic lead in uraninite ordinarily runs from about 3 to 10 weight per cent PbO, but as much as 19 per cent PbO has been reported. Curite, with the highest content of Pb of the secondary hydrated uranyl oxides, is very rare in gum-

mite alteration pseudomorphs. Schoepite and becquerelite, with little or no lead, also are very rare. At Katanga, its principal locality, curite is formed chiefly by direct crystallization from lead-containing solutions and is relatively late-formed in the paragenesis. The question of the valence state of the Pb present in uraninite and in some of the red-colored secondary oxides requires further investigation. It always is reported as Pb^2 by analysts. Wasserstein (1954), in his work on the determination of age by unit-cell measurements of uraninite, assumes the lead to be present as Pb^4 .

In the third stage of alteration, the hydrated oxides of the gummite zone react chemically with traversing solutions, ordinarily of the meteoric circulation but possibly at times of hydrothermal origin, with the formation of uranyl silicates and, occasionally, of the uranyl carbonate rutherfordine. Calcium ordinarily is added at this time, in addition to silica and water, giving rise to the characteristic minerals of this zone, uranophane, beta-uranophane, and Mineral *B*. Soddyite, a uranyl silicate lacking Pb, Ca, or other cations, also may form at this stage but is rare. Lead, retained during the gummite stage, ordinarily is completely lost during the formation of the silicates. Sometimes it is retained and concentrated relative to uranium with the formation of kasolite, with 37.5 per cent PbO , and Pb also is sometimes present in solid solution in Mineral *B*. Uranium, present in the silicates of this stage in amounts between 49 and 67 per cent UO_3 , apparently must be lost in part since the alteration commonly takes place without appreciable change in volume (pseudomorphic).

The history of the thorium and the rare earths contained in the original uraninite is not entirely clear. Analyses 4, 9, 12, and 15 to 19 of Table 1 indicate that these elements are retained at least in part in the minerals of the gummite zone. This also is indicated by a spectrographic study of zoned pseudomorphs made for the writer by Mr. H. C. Harrison. This study further showed that the thorium and rare earths tend to be leached out during the silicate stage of alteration. An analysis of uranophane cited beyond, however, indicates that thorium and rare earths may be retained in small part in the silicate stage, but these elements are not found in most analyses of uranophane and beta-uranophane. The studies made of changes in the ratios of uranium, thorium, and lead during the oxidation of uraninite, in connection with the determination of geologic age by the U, Th/Pb method, indicate that thorium tends to decrease and lead to increase relative to uranium as alteration proceeds. [See Bakken, Gleditsch, and Pappas (1948), Gleditsch and Bakken (1937), Bakken and Gleditsch (1938), Alter and Kipp (1937), Hecht (1931), and Hecht and Kroupa (1936).] These observations apply to uraninite in the

initial stages of alteration. Uraninite that has passed into the gummite stage of alteration is worthless for age determination work. Føyn (1937) has described experiments on the leaching of uraninite in water in closed tubes at 190° C. Yagoda (1945) describes the occurrence of radiocolloid aggregates in gummite and gives a color photograph of a gummite specimen. The radiocolloids apparently owe their origin to the leaching and local reprecipitation of radiogenic radium as radium sulfate, which has since largely decayed to $Pb^{206}SO_4$.

Clarkeite contains sodium and calcium as essential constituents, together with lead, and occurs in the inner part of some uraninite pseudomorphs in pegmatites. The writer inclines to the view of Ross, Henderson, and Posnjak (1931) that the clarkeite was formed by reaction of the uraninite with late pegmatitic hydrothermal solutions containing alkalis. The outer gummite and silicate zones of these pseudomorphs probably were formed much later by meteoric solutions.

DESCRIPTION OF MINERALS PRESENT IN GUMMITE

Becquerelite, $7UO_3 \cdot 11H_2O$ (?)

Becquerelite was not identified as a constituent of the gummite specimens examined in the present study. It has been reported by Steinkuhler (1923) and Hacquaert (1927) in uraninite pseudomorphs from Katanga. The mineral ordinarily occurs at this locality not as an alteration product but as drusy crusts and aggregates that have been deposited directly from solution in the supergene zone. It was observed as a direct alteration product of massive uraninite, however, in specimens from the Monument No. 2 mine, Apache County, Arizona, the Delta mine, San Rafael Swell, Utah, and Lake Athabaska, Canada. The rarity of becquerelite in gummite is rather surprising, especially since the mineral apparently can contain lead in solid solution. Schoep (1936) describes specimens from Katanga showing a central core of uraninite that has altered first to becquerelite and orange fourmarierite and later to green vandenbrandeite and cuprosklodowskite. Scheerer (1847) described a uraninite crystal from a pegmatite in Norway that had been altered to a pale yellow hydrated uranyl oxide, possibly becquerelite or schoepite.

Beta-uranophane, $Ca(UO_2)_2(SiO_3)_2(OH)_2 \cdot 5H_2O$

Beta-uranophane, the monoclinic polymorph of uranophane, was found in a few occurrences as the chief constituent of the outer, silicate zone of alteration. In these occurrences the mineral is identical in color and general appearance with uranophane (which see); it is best distinguished by optical and x -ray tests. Beta-uranophane has inclined extinction and relatively high indices of refraction (Table 3). Steinocher and

Nováček (1939) note that beta-uranophane may convert to uranophane when finely crushed, so that care must be taken in preparing samples for x -ray or optical study. In gummite pseudomorphs, beta-uranophane characteristically has a yellowish-green color sometimes inclining to lemon yellow. Dense material is slightly translucent and has a weak vitreous to waxy-vitreous luster. Like uranophane, the mineral also occurs as soft, rather porous aggregates of a dull or earthy luster and then inclines to straw yellow and brownish-yellow shades of color. This soft material seems to be a product of leaching of the dense, vitreous pseudomorphs. Under the microscope the mineral appears as matted aggregates of fibers, often minute in size and distorted, and accurate optical measurements are difficult to obtain. Beta-uranophane was identified in material from the following localities: various pegmatites in the Spruce Pine district, Mitchell County, North Carolina; Newry, Maine; the Ruggles pegmatite, Grafton Center, and the Palermo pegmatite, North Groton, New Hampshire; Easton, Pennsylvania; Rajputana, India. It was also found as crusts of acicular crystals associated with altering uraninite at Theano Point, Lake Superior, Canada, and Wölsendorf, Bavaria.

Clarkeite, $(\text{Na}, \text{Ca}, \text{Pb}, \text{H}_2\text{O})_2\text{U}_2(\text{O}, \text{H}_2\text{O})_7$

Clarkeite was identified in a number of specimens from the Spruce Pine pegmatite district, North Carolina—the type locality for the mineral. It also was found at a new locality, the Ajmer district, Rajputana, India. The composition and properties of clarkite have been recently described by Gruner (1954) and by Frondel and Meyrowitz (1956). Clarkeite occurs as a direct alteration of uraninite. It is earlier formed than the minerals of the orange-red gummite zone and may alter to them. It differs from these minerals in its dark-brown or mahogany-brown color, and in containing Na and Ca in addition to Pb. It occurs as dense, fine-grained aggregates. The indices of refraction are high, near those of curite, and vary considerably with variation in the ratio of Na, Ca, and Pb (Table 3). A positive identification is best made by x -ray means (Table 4). The mineral also resembles the brownish-black, relatively highly oxidized types of uraninite. Clarkeite has been said to occur in reddish-brown gummite at the Ruggles pegmatite, Grafton Center, New Hampshire, but it was not observed in specimens from this locality examined in the present study.

Curite, $3\text{PbO} \cdot 8\text{UO}_3 \cdot 4\text{H}_2\text{O}(?)$

Curite was not identified with certainty in any of the gummite pseudomorphs here examined. It may have been admixed in very small amounts

in some of the fourmarierite pseudomorphs. In one instance an x -ray pattern very close to that of curite was obtained from a deep orange-red gummite from Mitchell County, North Carolina, but the indices of refraction were much too low to confirm the identification. The orange-red to red color of many gummite specimens is not due to curite, as has been suspected in the past, but chiefly to fourmarierite and in part to vandendriesscheite. Curite pseudomorphs after uraninite crystals from Katanga have been described by Vernadsky and Chamie (1924), Van Aubel (1927), Schoep (1930), and Hacquaert (1927). Schoep and De Leenheer (1937) found that certain colloform crusts from Katanga, termed gummite by them, were composed of curite (analysis 10, Table 1). Their belief that all gummite is composed of curite is incorrect. A transparent, isotropic substance with a bright-scarlet color found here as an alteration product of uraninite from Fone, Gjerstad, Norway, may be an amorphous gel form of curite. It has an index of refraction slightly over 2, and did not give an x -ray diffraction pattern. A flame-scarlet alteration product of uraninite from Villeneuve, Quebec, analyzed by Ellsworth (1930) and cited in Table 1, might contain curite.

Fourmarierite, $\text{PbO} \cdot 4\text{UO}_3 \cdot 7\text{H}_2\text{O}$ (?)

Chemically, it is known from the reported analyses (Table 1) and from numerous spectrographic analyses made during the present study, that the minerals of the gummite zone of alteration are essentially hydrated oxides of uranium and lead. Barium, calcium, thorium, and alkalis often are also present in significant amounts in addition to lead. For practical purposes of identification, the minerals that fall into this general region of composition may be divided into two groups. The first group, comprising curite, clarkeite, becquerelite, billietite, and Mineral A, are relatively easy to identify in the present connection because their x -ray patterns are distinctive and their optical and other properties are also diagnostic. The second group, comprising fourmarierite, vandendriesscheite, schoepite, Mineral C, masuyite, and richetite, are difficult to identify. The x -ray powder patterns of all of these substances are very similar, and in the case of faint or diffuse patterns or of mutual admixture a positive discrimination becomes difficult or impossible by this method. In this case, the optical properties are of aid: schoepite without lead has much lower indices of refraction than the others, with $\gamma = 1.74$; vandendriesscheite and Mineral C have indices usually in the range 1.76–1.85; fourmarierite has higher indices, between 1.84 and 1.96 for the most part; the problematic mineral richetite is said to have β and γ from 2.00 to 2.07; masuyite has α 1.785, β 1.906, γ 1.917 according to one description and β 2.11, γ 2.15 according to another. Complete optical descriptions

are given in Table 3. In the present study, the standard *x*-ray patterns employed were obtained from type analyzed specimens of these minerals (with the exception of richetite, for which data and specimens are lacking). The *x*-ray pattern of masuyite is virtually identical with that of vandendriesscheite. The identification of these minerals in gummite was regarded as certain only if both the optical properties and the *x*-ray pattern answered the standard description. If, as was often the case, the optical properties were divergent or the optical properties answered the description and the *x*-ray pattern was diffuse and ambiguous, the identification was regarded as tentative. This procedure is not wholly satisfactory, as there are many uncertainties about the mutual relations of the minerals here classed together. Thus it is an open question whether the composition of these minerals is fixed, or whether there is a partial or complete variation in composition in point of lead content, with attendant variation in the *x*-ray pattern or optics, between some or all of them. Further, it is not known how the presence of barium or calcium in solid solution for lead affects the *x*-ray and optical constants. This is an important factor in the case of fourmarierite and vandendriesscheite at least. The barium and calcium analogues of fourmarierite have been synthesized.¹ They are isostructural with fourmarierite, and complete series probably extend between all three members. The barium uranyl oxide billietite was not identified in gummite, but it was observed as sharp crystals coating cracks in a fourmarierite pseudomorph after uraninite from Katanga. In the latter specimen the complete sequence of deposition was uraninite (core), fourmarierite, billietite, uranophane, torbernite.

Fourmarierite was identified in gummite from the following localities: Wölsendorf, Bavaria; Tvedestrand, Norway; as an alteration of pitchblende from an unstated locality in Bohemia, probably Joachimsthal; Bisundi, Rajputana, India; the Spruce Pine district, Mitchell County, North Carolina; Newry, Maine; the Katanga district, Belgian Congo; Mica Lakes area, Hahns Peak, Colorado. It also was found as thin colloform crusts on altering uraninite from Great Bear Lake, Canada. Some of the material from these localities had indices of refraction slightly higher than those reported in the literature, with γ 1.95–1.97. Fourmarierite was doubtfully identified as a minor constituent of gummite from a number of localities, including Morogoro, Tanganyika; Rajputana, India; the Palermo and Ruggles pegmatites in New Hampshire; Pamplonita, Colombia (with γ a little over 2); the Ingersoll mine, Pennington County, South Dakota; Bathurst, Lanark County, Ontario;

¹ By Miss Eleanor R. Berman, Department of Mineralogy, Harvard University, 1954.

Wilberforce, Cardiff Township, Ontario. The mineral occurs as dense, microcrystalline aggregates, and the color is orange red. The color is lighter than that of curite. There is no question but that the indices of refraction of the mineral vary by at least 0.04 due to variation in composition, presumably the substitution of Ba and Ca for Pb.

Sarkar and Sen (1946) made an *x*-ray powder study of an orange-yellow gummite from Rajputana, India, which encased a residual of uraninite and was in turn surrounded by lemon-yellow uranophane. They concluded that the mineral was gummite (which they accept as a species name) from the near agreement of their *d*-spacings with the data given by Ross, Henderson, and Posnjak (1931) for an unanalyzed specimen of gummite (in the broad sense) from Mitchell County, North Carolina. This pattern is of the fourmarierite type but cannot be more closely identified. The optical properties given by Ross, Henderson, and Posnjak (α 1.742, β 1.762, γ 1.776, $2V$ 60°, $r < v$) differ from those of fourmarierite and vandendriesscheite but may represent Mineral C. Clarkeite and fourmarierite(?) were recognized in gummite specimens from Rajputana during the present study. Two partial analyses of impure gummite from Rajputana cited by Krishnan (1948) show 8.26 and 19.10 per cent PbO. Crookshank (1948) describes some of the minerals from the Rajputana pegmatites.

Kasolite, $\text{Pb}(\text{UO}_2)(\text{SiO}_3)(\text{OH})_2(?)$

Kasolite occurs in the silicate zone of alteration as earthy to firm microcrystalline aggregates of minute shreds and fibers. Distinct euhedral crystals such as characterize the Katanga material were not observed. The mineral has a brownish-yellow to pale-brown color, and closely resembles the isostructural species Mineral B (which see). It often occurs more or less admixed with shreds and fibers of uranophane. Kasolite was identified as the principal or sole constituent of the outer silicate zone in uraninite pseudomorphs from Kakanas and Gordonias, Africa; the Ruggles pegmatite, Grafton Center, New Hampshire; and Bisundi, Rajputana, India. It also was observed as a very minor constituent of the silicate zone in material from the Morogoro district, Tanganyika, and a few other localities. At the Ruggles pegmatite, kasolite occurs both as a thin outer crust about the orange gummite pseudomorphs for which the locality is noted and as complete pseudomorphs of a rather porous consistency.

Phosphuranylite, $\text{Ca}(\text{UO}_2)_4(\text{PO}_4)_2(\text{OH})_4 \cdot 7\text{H}_2\text{O}(?)$

Phosphuranylite was observed admixed in small amounts with uranophane in the surface parts of altered uraninite crystals from Spruce Pine,

Mitchell County, North Carolina, and the Ruggles pegmatite, Grafton Center, New Hampshire. Cubical molds of uraninite crystals, some empty and others more or less completely filled with golden-yellow, earthy phosphuranylite were noted in specimens from the Flat Rock mine, Mitchell County, North Carolina; these pseudomorphs, however, appear to be due to the infilling of empty cavities (casts) and not to the alteration of uraninite in place. Phosphuranylite occurs much more commonly as films and coatings on fracture surfaces in the immediate neighborhood of uraninite pseudomorphs in pegmatites (Fron del, 1950). An occurrence of phosphuranylite(?) with gummite and uraninite in eastern Kuangsi province, China, is described by Nan and Wu (1943).

Rutherfordine, $(\text{UO}_2)(\text{CO}_3)$

Earthy, yellow pseudomorphs of rutherfordine after uraninite crystals were described by Marckwald (1906) from a pegmatite in the Morogoro district, Tanganyika, Africa. Specimens examined from this locality showed a central core of uraninite, a surrounding zone of orange-red gummite, and a pulverulent outer zone of rutherfordine usually much admixed with uranophane and kasolite. The mineral occurs similarly in pegmatite at Newry, Maine, and Beryl Mountain, New Hampshire. A description of the mineral from its known localities is given by Fron del and Meyrowitz (1956). Rutherfordine effervesces strongly in dilute acids, although the reaction starts slowly, and this provides a useful aid in its recognition. The mineral often is much too fine grained for satisfactory optical study, and an *x*-ray powder pattern may be required. The uranyl carbonates sharpite and studdite and the carbonates of uranium containing additional cations, such as liebigite, were not encountered in this study. In its known occurrences rutherfordine appears to be produced by the action of carbonated surface waters on the hydrated oxides of the gummite zone.

Schoepite, $2\text{UO}_3 \cdot 5\text{H}_2\text{O}(?)$

Only one example of schoepite was observed in the gummite zone of alteration. It occurs as golden-yellow to yellow, earthy pseudomorphs after uraninite in the Beryl Mountain pegmatite, near Acworth, New Hampshire. Fine-grained Mineral *B*, uranophane, and rutherfordine were associated. At Katanga, the principal locality for the mineral, schoepite is a drusy or granular massive deposit from solutions and is not a pseudomorphic alteration product. Hacquaert (1927), however, has noted its presence with becquerelite in a uraninite pseudomorph from this locality. Its rarity in gummite is due to the general presence of lead, which leads to the formation of hydrated oxides such as vanden-

driesscheite and fourmarierite. Schoepite also was observed as a yellow crust on massive uraninite from Hottah Lake, Canada, and from the Delta mine, San Rafael Swell, Utah.

Sklodowskite, $\text{Mg}(\text{UO}_2)_2(\text{SiO}_3)_2(\text{OH})_2 \cdot 5\text{H}_2\text{O}$

Sklodowskite was not identified in the present study among the uranium silicates comprising the outer zone of alteration. It has been observed, however, by Hacquaert (1927) with curite and cerussite comprising pseudomorphs after uraninite from Kasolo, Katanga.

Soddyite, $(\text{UO}_2)_6(\text{SiO}_4)_2(\text{OH})_2 \cdot 5\text{H}_2\text{O}$

Soddyite was identified only once in the present study, although its presence in very small amounts admixed with uranophane was suspected in material from a number of localities. It occurs abundantly at the Ruggles pegmatite, Grafton Center, New Hampshire, as complete pseudomorphs after uraninite. These are yellow to straw-yellow in color, with a dense to almost pulverulent consistency, and are composed of minute shreds and fibers of soddyite admixed with small amounts of uranophane. In the hand specimen they resembled very closely the earthy types of uranophane and Mineral B pseudomorphs from this and other localities. Soddyite also has been found in the Norrabees pegmatite in Namaqualand, South Africa (Gevers, Patridge, and Joubert, 1937), where it presumably is an alteration product of uraninite. Soddyite may be present in a gummite pseudomorph from Alto do Tibiri, Paraiba, Brazil, analyzed by Florencio (1951) (column 14, Table 1). Microcrystalline soddyite is rather similar optically to beta-uranophane and phosphuranylite, although it lacks the distinct golden pleochroism of the latter, and is best identified by *x*-ray methods.

Uranophane, $\text{Ca}(\text{UO}_2)_2(\text{SiO}_3)_2(\text{OH})_2 \cdot 5\text{H}_2\text{O}$

The occurrence of a calcium uranyl silicate in the outermost, greenish zone of alteration of uraninite pseudomorphs, bordering on and veining the orange-red gummite zone, was established chemically by Genth (1879) and Von Foullon (1883) in material from pegmatites in Mitchell County, North Carolina. A probably identical mineral occurring as an alteration of uraninite from Arendal, Norway, was analyzed by Norden-skiöld (1884). The existence of uranophane in the siliceous outer zone in such pseudomorphs also was noted by Hidden (1881), Tipper (1919), Hacquaert (1927), Ross, Henderson and Posnjak (1931), Ellsworth and Osborne (1934), Buttgenbach (1922), Schoep (1921), Sarkar and Sen (1946), and others. The first specimen sent to Belgium after the discovery of uranium in Katanga in 1913 was described by Buttgenbach (1926) as

pitchblende altered to gummite and uranophane. This mineral has been called uranophane (or uranotil) in the older literature, but it was shown by Nováček (1935) and Steinocher and Nováček (1939) that the compound is dimorphous, orthorhombic (uranophane) and monoclinic (beta-uranophane). The distinction between these polymorphs rests chiefly on optical and *x*-ray criteria.

There is also the question of the identity of uranophane and uranotil. Both names are found in some modern literature, as distinct species, based on the belief that the acicular calcium uranyl silicate from Wölsendorf, Bavaria, described by Bořický (1870) under the name uranotil differs in composition from the very similar mineral described earlier from Kupferberg, Silesia, by Websky (1853) as uranophane. Websky's analysis unfortunately was made on very impure material. An *x*-ray and optical study made here of uranophane from Kupferberg and of uranotil from Wölsendorf has shown conclusively that these two minerals are identical. The name uranophane (and beta-uranophane) should be employed for the species, as was already concluded by Dana (1892).

The uranophane of the silicate zone of alteration generally appears as a hard, dense mass, without structure visible to the unaided eye, and has a waxy to subvitreous luster. The color of the mineral commonly is a pale greenish yellow, but varies to yellowish green, dull green, yellow, orange yellow, and straw yellow. Occasionally it forms relatively soft to porous masses with a dull to earthy luster and a straw-yellow to brownish-yellow color. Under the microscope, the mineral is microcrystalline and composed of shreds or fibers in a confused or matted array. There is almost always at least a small degree of admixture with other minerals. The indices of refraction may be difficult to obtain accurately, because of small particle size. Beta-uranophane, which otherwise resembles the present species very closely, has higher indices and the extinction is inclined (Table 3). The *x*-ray patterns of the two minerals are quite different (Table 4). Uranophane (and beta-uranophane) ordinarily are separated from the central core of uraninite pseudomorphs by an intermediate zone of gummite, from which the silicates have been derived. In a few instances, however, uranophane was observed forming directly from uraninite.

Uranophane was identified as the chief constituent of the silicate zone in specimens from a large number of localities. These included numerous places in the pegmatite districts of North Carolina, New Hampshire, Maine, Connecticut, Colorado, and the Black Hills, South Dakota. Among the foreign localities may be mentioned Calamuchita, Córdoba, Argentina; Rajputana, India; Karelia, Russia; Katanga, Belgian Congo; Morogoro, Tanganyika. The mineral also was identified as fibrous crusts

and aggregates associated with altering uraninite, but not pseudomorphous after it, from Lake Athabaska, Saskatchewan, and Theano Point, Lake Superior, Canada; Joachimsthal, Bohemia; Katanga district, Belgian Congo; Wölsendorf, Bavaria; the Grants area, New Mexico; the Marysvale and Henry Mountains districts, Utah.

A specimen from Mitchell County, North Carolina, showed a uraninite cube that had been wholly altered to a soft, porous, straw-yellow aggregate of uranophane. A chemical analysis of this material by F. A. Gonyer, cited below, is of interest in showing the presence of a considerable amount of thorium and yttrium in substitution for calcium. A semi-

CaO	ThO ₂ and Y ₂ O ₃	UO ₃	SiO ₂	H ₂ O	Total
5.77	2.60	66.40	13.40	11.71	99.88

quantitative spectrographic analysis by H. C. Harrison showed that Y composed about 10 per cent of the mixed (Th, Y) oxides. Optically, this material had $\alpha \sim 1.642$ (nearly colorless), $\gamma \sim 1.668$ (pale yellow) in Na light, with Z parallel to the elongation.

Vandendriesscheite, $\text{PbO} \cdot 7\text{UO}_3 \cdot 12\text{H}_2\text{O}(\?)$

The problems attending the identification of vandendriesscheite have been discussed earlier under fourmarierite, which see. Vandendriesscheite was identified in gummite pseudomorphs from the following localities: numerous places in the Spruce Pine district, Mitchell County, North Carolina, including the Wiseman, McKinney, Deak and various unstated mines in this district; Newry, Maine; the Palermo and Ruggles pegmatites in New Hampshire; numerous specimens from the Katanga district, Belgian Congo; Joachimsthal, Bohemia. The mineral also occurs as crusts and minute crystals (the "Mineral X" of Palache and Berman, 1933) on altering uraninite at Great Bear Lake, Canada. It was observed as their colloform films on uraninite from Hottah Lake, N.W.T., Canada. Vandendriesscheite was doubtfully identified from a number of other localities, and the mineral appears to be more common than fourmarierite. It often occurs admixed with other oxidic minerals of both higher and lower indices, and uranophane is commonly present in small amounts. Vandendriesscheite occurs as dense microcrystalline aggregates. The color varies from yellow-orange, brownish orange, and orange to orange-red. The reddish shades are less typical than with fourmarierite. Usually only the mean index of refraction can be measured due to small particle size; this varies from about 1.77 to 1.82. Some material doubtfully identified as vandendriesscheite had α as low as about 1.71-1.74. There seems to be little doubt that the indices of refraction of this min-

eral vary over at least 0.04 due to compositional variation, presumably a substitution of Ba or Ca for Pb.

Unidentified Mineral A

This mineral was recognized in the gummite zone of specimens from six localities: pegmatites at North Wilton, Palermo, Ruggles, Alstead, and Beryl Mountain, all in New Hampshire, and a pegmatite at Grassy Creek, Mitchell County, North Carolina. The latter specimen was one of those described and analyzed by Von Fouchon (1883) in his study of gummite. Mineral *A* is particularly abundant at the Ruggles locality, where it is one of the chief constituents of the orange gummite pseudomorphs after dendritic aggregates of uraninite crystals for which the locality is noted (see Shaub (1938)). The mineral was recognized as a distinct and presumably new species on the basis of its *x*-ray powder pattern (Table 4). It occurs as microcrystalline aggregates of a bright-orange to orange-yellow or golden-yellow color. The reddish shades of color seen in many fourmarierite and some vandendriesscheite specimens are lacking. The luster varies from dull and earthy to weakly vitreous in hard, translucent aggregates. Under the microscope the mineral is yellow in color, not perceptibly pleochroic, and has low to moderate birefringence. Only the mean index of refraction could be obtained. This varies between 1.78 and 1.88 in most specimens, but one specimen from the Ruggles pegmatite had $n \sim 1.96$. Semiquantitative spectrographic analyses show that the mineral is an oxide of uranium or a uranate containing Pb, K, Na, Ca, and Ba in significant amounts. The Ruggles material with $n \sim 1.96$ contained Pb between 5 and 10 per cent, K and Ca 0.5–1 per cent, Na and Ba 0.1–0.5 per cent. Material from Palermo with $n \sim 1.84$ contained both Pb and K in the range 1 to 5 per cent, and Ca, Ba and Na in the range 0.1 to 0.5 per cent. Possibly several different isostructural minerals varying in the mutual substitution of Pb, K, Ca, etc., with accompanying variation in the indices of refraction, are represented. The *x*-ray patterns of all specimens are virtually identical. There was a complete change in the *x*-ray pattern of the Ruggles material after heating to 500° C. Study of this mineral is continuing.

Unidentified Mineral B

Mineral *B*, apparently the calcium analogue of kasolite, is a relatively common constituent of the silicate zone of alteration. It was first recognized as a probable new species during the present study, and a detailed description is in the course of preparation. Mineral *B* occurs as dense to earthy microcrystalline aggregates composed of minute shreds and fibers. Its color is straw yellow to pale yellowish brown, and lighter than

typical kasolite. The x -ray powder pattern is virtually identical with that of kasolite, but the indices of refraction are considerably less (Table 3). The presence of calcium as a major constituent was established by chemical tests. The mineral often is so fine grained that only the mean index of refraction can be obtained; this usually ranges between 1.81 and 1.84. Admixture with uranophane is common. Mineral *B* occurs both as complete pseudomorphs and as zones surrounding orange-red gummite; it also has been observed as earthy crusts along fracture surfaces in the matrix. The mineral was recognized from the following localities: the Bob Ingersoll pegmatite, Pennington County, South Dakota; the Rugles pegmatite near Grafton Center, and the Beryl Mountain pegmatite near Acworth, both in New Hampshire; the Mica Lake area, Hahn's Peak, Colorado; and Bisundi, Rajputana, India. It was doubtfully identified in material from Karelia, Russia, and several other localities.

Unidentified Mineral C

This apparently new species was recognized on the basis of its x -ray powder pattern (Table 4). The pattern closely resembles but is distinct from the patterns of fourmarierite and schoepite. The pattern seems to be definite and characteristic as it was given by natural material from five different localities and also by a sample of synthetic hydrated lead diuranate. Mineral *C* occurs in the gummite zone of alteration as dense, microcrystalline aggregates with an orange-brown, reddish-brown or chocolate-brown color. The powder is mustard yellow. The color differs from the orange-red color of fourmarierite and some vandendriesscheite, and the yellow to orange color of Mineral *A*. Clarkeite, however, has a very similar color. The mineral was observed from the following localities: Mitchell County, North Carolina, at the Wiseman mine (analysis 4, Table 1), the McKinney mine, and at an unstated locality (analysis 12, Table 1); Easton, Pennsylvania, where it is an alteration of uraninite or uranoan thorianite; Newry, Maine. The material from the Wiseman mine is that described by Ross, Henderson and Posnjak (1931). The optics given by these authors for "gummite" do not refer to the present mineral; the material of their analysis sample, here examined, has indices between 1.77 and 1.82. A similar range is shown by the material from the other localities stated, with the exception of the material of analysis 12, Table 1, from Mitchell County, which has higher indices, 1.85-1.89. The indices are considerably lower than those of fourmarierite and clarkite, but comparable to those of vandendriesscheite. Schoepite has much lower indices of refraction. The fine-grained nature of the material makes optical study difficult. The two available analyses of this mineral, cited in Table 1, were made on slightly inhomogeneous material.

A semiquantitative spectrographic analysis of the Easton material showed U as the major constituent with Th, Pb, Ca, and Ba present in amounts between 1 and 10 per cent. Mineral *C* apparently is related structurally to fourmarierite. A further description of this mineral with additional analyses is planned.

ACKNOWLEDGMENTS

The present study was done by the U. S. Geological Survey on behalf of the Division of Raw Materials of the U. S. Atomic Energy Commission. The writer wishes to express his appreciation to Mrs. Judith W. Frondel for supplying new *x*-ray and optical data on vandendriesscheite and masuyite, and to Miss Mary Mrose, U. S. Geological Survey, for aiding in the preparation of *x*-ray powder photographs. Mr. H. C. Harrison, Rhode Island State College, kindly made a number of qualitative spectrographic analyses. Additional semiquantitative spectrographic analyses were obtained through Miss K. E. Valentine and Mr. Charles Annell, U. S. Geological Survey. A number of specimens were borrowed for study from the U. S. National Museum through the courtesy of Dr. George Switzer. Thanks also are expressed to various members of the Boston Mineral Club, particularly to Mr. Rudolph Bartsch, for their interest in supplying for study specimens of uranium minerals from pegmatite localities in New England. Mr. H. A. Ashley of North Rumney, New Hampshire, generously gave a collection of uranium minerals from the Palermo and Beryl Mountain pegmatites and facilitated collecting at those localities. Gummite specimens also were obtained for study from the Royal Ontario Museum through the courtesy of Dr. V. Ben Meen.

TABLE 3. OPTICAL PROPERTIES OF MINERALS PRESENT IN GUMMITE

Name	Symmetry	Indices and pleochroism			Sign, 2V, Disp.	Remarks
		α	β	γ		
Becquerelite	Orthorhombic	1.72-1.75 pale yellow	1.81-1.82 deep yellow	1.82-1.83 deep yellow	(-), 30° $r > v$	X perp. cleavage.
Beta-uranophane	Monoclinic	1.66-1.67 colorless	1.68-1.69 lemon- yellow	1.69-1.71 lemon- yellow	(-), 40°-70°, $r > v$	X = b , Z \setminus elong. = 18°-57°. Fi- bers.
Clarkeite	Orthorhombic?	2.00	2.10	2.11	(-), mod. $r < v$	Indices vary; some with mean index 1.94-1.97
Curite	Orthorhombic	2.05-2.06 pale yellow	2.07-2.11 light red- orange	2.11-2.15 dark red- orange	(-), large $r > v$	Z = elong.
Fourmarierite	Orthorhombic	1.85-1.86 colorless	1.90-1.92 pale yellow to orange	1.90-1.94 yellow to orange	(-), mod. to large $r > v$	X perp. cleav., Y = elong.
Kasolite	Monoclinic	1.88-1.90	1.90-1.91	1.94-1.96	(+), 42°	X = elong. Faint yellow pleochro- ism. Fibers.
Masuyite	Orthorhombic	1.785 pale yellow	1.906 deep golden	1.917 deep golden	(-), 50°, $r > v$	X perp. cleav. Also with indices 2.11-2.15 (?).
Meta-autunite	Tetragonal	1.58-1.60 pale yellow	1.59-1.61 yellow	1.59-1.61 yellow	(-), 0° to small, $r > v$	X perp. plates and cleavage. In- dices and 2V vary with H ₂ O.
Mineral A	Not known	Mean index 1.78-1.88, variable			(+) mod.	Yellow. Not pleo- chroic (?). Also with $n = 1.96$.
Mineral B	Monoclinic	1.825	~1.84	1.867	(+) mod.	Mean index 1.81- 1.83 when fine grained. Faint pleochroism.
Mineral C	Not known	Mean index 1.77-1.82, varia- ble, in part higher?			(+) mod.	Orange-brown. Not pleochroic ?
Phosphuranylite	Orthorhombic	1.66-1.67 pale yellow	1.70-1.72 golden yellow	1.70-1.72 golden yellow	(-), 5°-25°, $r > v$	Z perp. plates. Indices increase with Pb content.
Rutherfordine	Orthorhombic	1.721 pale yellow	1.729 golden yellow	1.757 greenish yellow	(+), large	Y = elong.
Schoepite	Orthorhombic	1.69-1.70 colorless	1.71-1.72 lemon- yellow	1.73-1.74 lemon- yellow	(-), large $r > v$	X perp. cleav- age.
Sklodowskite	Orthorhombic	1.613	1.635	1.657	(-), large	Y = elong.
Soddyite	Orthorhombic	1.65 colorless	1.68 pale yellow	1.71 pale green- yellow	(-), large $r > v$	Often cloudy.
Uranophane	Orthorhombic	1.64 colorless	1.66 golden yellow	1.67 golden yellow	(-), small $r > v$	Z = elong. Paral- lel.
Vandriesscheite	Orthorhombic	1.76-1.79 colorless	1.81-1.84 golden yellow	1.82-1.85 golden yellow	(-), med. $r > v$	X perp. cleavage. Mean index 1.77- 1.82.

TABLE 4. X-RAY POWDER DATA FOR MINERALS IN GUMMITE

Copper radiation, nickel filter, in Ångstroms

Note: In most instances only the inner lines of the patterns are cited, sufficient to ensure discrimination.

Becquerelite		Beta-uranophane		Clarkeite		Curite	
<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>
1	8.51	10	7.83	1	10.98	10	6.28
10	7.50	4	6.66	1	8.33	1	4.80
2	6.63	4	6.10	2	6.70	4	4.55
1	6.24	4	5.07	8	5.77	1	4.17
1	5.63	4	4.85	4	4.09	9	3.97
6	4.71	4	4.55	9	3.34	4	3.53
1	4.31	3	4.11	10	3.17	4	3.36
8	3.75	9	3.90	3	2.92	8	3.14
8	3.56	1	3.75	5	2.69	4	3.07
2	3.48	6	3.51	4	2.61	2	2.93
2	3.39	1	3.41	3	2.45	1	2.81
9	3.22	2	3.35	2	2.06	4	2.70
1	3.13	5	3.19	7	1.968		
2	2.97	4	3.04	6	1.863		
		3	2.99				
		2	2.91				
		4	2.83				
		3	2.78				
		5	2.59				

Fourmarierite		Kasolite		Phosphuranylite		Rutherfordine	
<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>
2	7.86	6	6.61	1	10.16	10	4.60
4	6.83	2	6.19	10	7.83	8	4.27
9	3.45	4	5.31	1/2	7.20	8	3.87
10	3.09	2	4.76	1	6.33	9	3.20
4	2.73	8b	4.19	8	5.83	5	2.62
6	2.44	7	3.53	1/2	5.37	2	2.48
5	2.26	1	3.38	1	4.96	3	2.41
2	2.15	10	3.26	2	4.72	4	2.28
2	2.09	5	3.07	3	4.33	6	2.14
6	1.996	9	2.93	9	3.97	8	2.04
8	1.907	3	2.73	1	3.83	1	1.94
2	1.817	3	2.64	1	3.44	3	1.91
				2	3.36	2	1.87
				6	3.12	2	1.73
				4	3.07		
				2	2.94		
				6	2.86		

b=broad line.

TABLE 4. X-RAY POWDER DATA FOR MINERALS IN GUMMITE—*Continued*

Schoepite		Sklodowskite		Soddyite		Uranophane	
<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>
10	7.49	10	8.42	8	6.14	10	7.88
1	6.74	2	6.37	3	4.71	4	6.61
2	6.09	5	5.91	9	4.48	4	5.42
2	4.08	4	4.82	2	3.73	5	4.76
8	3.64	1	4.51	10	3.32	2	4.29
9	3.26	4	4.33	2	3.22	9	3.94
1	3.01	8	4.19	3	2.95	4	3.60
2	2.83	5	4.00	2	2.78	4	3.51
7	2.60	6	3.52	7	2.69	1	3.41
3	2.48	7	3.27	6	2.47	1	3.35
2	2.35	6	3.00	1	2.39	5	3.20
1	2.27	5	2.87	2	2.32	1	3.09
1	2.21	2	2.80	2	2.26	8	2.99
4	2.10	2	2.74	1	2.18	8	2.91
6	2.05	1	2.70	4	2.09		
1	1.981	1	2.66				

Vandriesscheite		Mineral A		Mineral C	
<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>
2	8.27	7	6.37	5	6.95
10	7.31	10	3.49	1	5.85
3	6.49	7b	3.18	1	4.12
1	4.85	5	3.02	10	3.46
2	4.39	3b	2.61	10	3.09
1	4.14	1	2.09	1	2.77
2	3.93	2	2.00	3	2.44
8	3.58	3b	1.92	2	2.29
1	3.37	1	1.81	5	1.991
9	3.19	3	1.75	6	1.916
1	3.00	1	1.57	6	1.730
3	2.73	1	1.52	4	1.673
5	2.53			2	1.548
2	2.40			1	1.509
1	2.32			2	1.383
2	2.05			1	1.355
7	1.989			2	1.305
3	1.908			2	1.283

b=broad line.

REFERENCES

- AHLFELD, F., AND ANGELLELI, V. (1948), Mineral species of the Argentine Republic: *Univ. Nac. Tucuman, Pub.* **458**.
- ALTER, C. M., AND KIPP, E. M. (1936), The variation of the lead-uranium-thorium ratio of a single crystal of Wilberforce, Ontario, uraninite: *Am. Jour. Sci.*, ser. 5, **32**, 120.
- AUBEL, R. VAN (1927), Sur la presence d'uraninite cristallisée. . . Katanga: *Comptes Rendus*, **185**, 586.
- BAIN, G. W. (1950), Geology of the fissionable materials: *Econ. Geology*, **45**, 273.
- BAKKEN, R., AND GLEDITSCH, E. (1938), Analyses in layers of a single crystal of cleveite, Auselmyren, Norway: *Am. Jour. Sci.*, ser. 5, **36**, 95.
- BAKKEN, R., GLEDITSCH, E., AND PAPPAS, A. C. (1948), The uraninites: *Soc. chim. France Bull.* (1948), 515.
- BOŘICKÝ, E. (1870), Mineralogische Mittheilungen I, Uranotil, ein neues Mineral von Wölsendorf in Bayern: *Gesell. Wiss. Prague Sitzungsber.*, 1870, 36.
- BREITHAUP, A. (1830), Übersicht des Mineral-Systems, Freiberg, p. 60.
- BREITHAUP, A. (1847), Handbuch der Mineralogie, Leipzig, p. 903.
- BROOKER, E. J., AND NUFFIELD, E. W. (1952), Studies of radioactive compounds IV. Pitchblende from Lake Athabaska: *Am. Mineral.*, **37**, 363.
- BUTTGENBACH, H. (1922), Note preliminaire sur des minerals d'uranium et de radium trouvés au Katanga: *Soc. géol. Belgique Annales, Pub. Congo belge*, **44**, C5.
- BUTTGENBACH, H. (1926), Mineralogie du Congo Belge: *Soc. royale Sci. Liege, Mém.*, ser. 3, **13**, 65.
- CHAUDHURI, B. D. N. (1944), On the radium content of a new mineral from Rajputana: *India Jour. Physics*, **18**, 257.
- CROOKSHANK, H. (1948), Minerals of the Rajputana pegmatites: *India Min. Geol. Met. Inst.*, **42**, 173.
- DANA, J. D. (1868), System of Mineralogy, 5th ed., New York, p. 179.
- DANA, E. S. (1892), System of Mineralogy, 6th ed., New York.
- ELLSWORTH, H. V. (1925), Radioactive minerals as geological age indicators: *Am. Jour. Sci.*, ser. 5, **9**, 127.
- ELLSWORTH, H. V. (1930), Four stages in the alteration of the Villeneuve uraninite: *Am. Mineral.*, **15**, 455.
- ELLSWORTH, H. V., AND OSBORNE, F. F. (1934), Uraninite from Lac Pied des Monts, Saguenay district, Quebec: *Am. Mineral.*, **19**, 421.
- EVERHART, D. L. (1951), Geology of uranium deposits: *U. S. Atomic Energy Comm.*, **RMO-732**.
- FLORENCIO, W. (1951), Cronogeologia dos pegmatitos Brasileiros: *Mineração e Met.*, **15**, 191.
- FOULLON, H. VON (1883), Über Verwitterungsprodukte des Uranpfecherzes: *Geol. Reichsanst. Wien, Jahrbuch* **33**.
- FØYN, E. (1937 and 1938), Über einige Verhältnisse in Uranmineralien: *Norsk geol. Tidsskr.*, **17**, 197 (1937); *Norske vidensk.-akad., Math.-nat. Kl. I*, no. 4 (1938).
- FREIESLEBEN, J. K. (1817), *Beitr. Min. Kenntnis Sachsen, Freiberg*, **2**, 187.
- FRONDEL, C. (1950), Studies on uranium minerals (V): Phosphuranylite: *Am. Mineral.*, **35**, 756.
- FRONDEL, C., AND MEYROWITZ, E. (1956), Studies on uranium minerals (XIX): Rutherfordine, diderichite, clarkeite: *Am. Mineral.*, **41**, 127.
- GENTH, F. A. (1879), Examination of the North Carolina uranium minerals: *Am. Chem. Soc. Jour.*, **1**, 89.

- GENTH, F. A. (1857), Contributions to mineralogy, no. 6, coracite: *Am. Jour. Sci.*, **23**, 421.
- GEORGE, D'A. (1949), Mineralogy of uranium and thorium bearing minerals: *U. S. Atomic Energy Comm.*, **RMO-563**.
- GEVERS, T., PARTRIDGE, F., AND JOUBERT, G. (1937), The pegmatite area south of the Orange River, Namaqualand: *South Africa Geol. Survey, mem.* **31**, 172.
- GLEDITSCH, E., AND BAKKEN, R. (1937), The determination of UO_2 and UO_3 in different layers of a single crystal of uraninite: *Mikrochimica Acta*, **1**, 83.
- GRUNER, J. W. (1954), The chemical formula of clarkeite: *Am. Mineral.*, **39**, 836-838.
- HACQUAERT, A. L. (1927), Over pseudomorphosen in radioactieve mineralen van Katanga: *Natuurk. tijdschr. Ned.-Indië*, **9**, 34.
- HADINGER, W. (1852 and 1853), Ueber den Eliasit von Joachimsthal: *Geol. Reichsanstalt Jahrbuch*, **3**, no. 4, p. 124 (1852); *Akad. Wiss. Wien Sitzungsber.*, **103** (1853).
- HAYDEN, H. H. (1914), General report for 1913. Pitchblende: *India Geol. Survey, Rec.* **44**, 24.
- HECHT, F. (1931), Zur Kritik der Altersbestimmung: *Akad. Wiss. Wien Sitzungsber.*, **140**, 2A, 599.
- HECHT, F., AND KROUPA, E. (1936), Die Bedeutung der quantitativen Microanalyse radioactiver Mineralien: *Zeits. anal. Chemie*, **106**, 82.
- HERMANN, R. (1859), Ueber die Zusammensetzung der zur Gruppe der Uransilicate gehörenden Mineralien; *Jour. prakt. Chemie*, **76**, 322.
- HEY, M. H. (1950), An index of mineral species and varieties, London.
- HIDDEN, W. E. (1881), Notes on some mineral localities in North Carolina: *Am. Jour. Sci.*, ser. 3, **22**, 22.
- HOFFMANN, G. C. (1901), On some new mineral occurrences in Canada: *Am. Jour. Sci.*, ser. 4, **11**, 149.
- KATZ, J. J., AND RABINOWITCH, E. (1951), The chemistry of uranium, Part I, New York, McGraw-Hill Book Co.
- KERR, W. C. (1877), Note on uranium minerals in North Carolina: *Am. Jour. Sci.*, ser. 3, **14**, 496.
- KERSTEN, C. (1832), Hyacinthrothes Pechuran von Johanngeorgenstadt: *Jour. für Chemie u. Physik*, **66**, 18.
- KHLOPIN, V. G. (1938), Oxygen method of determination of geological age: *Acad. sci. U.R.S.S. Bull., Math.-Nat. Cl.* 489.
- KOHL, E. (1954), Die metallischen Rohstoffe, **10**, Uran. Ferdinand Enke Verlag, Stuttgart.
- KRISHNAN, M. S. (1948), Uranium: *India Jour. Sci. Indus. Research*, **7A**, 30.
- LANG, A. H. (1952), Canadian deposits of uranium and thorium: *Canada Dept. Mines, Econ. Geol. Ser.*, **16**.
- LE CONTE, J. L. (1847), On coracite, a new ore of uranium: *Am. Jour. Sci.*, ser. 2, **3**, 173.
- MARCKWALD, W. (1906), Über Uranerze aus Deutsch-Ostafrika: *Centralbl. Mineralogie*, **761**.
- NAN, Y. T., AND WU, L. P. (1943), Note on some uranium minerals from eastern Kuangsi: *China Geol. Soc. Bull.*, **23**, 169.
- NORDENSKIÖLD, A. E. (1884), Yttrogummite: *Geol. fören. Stockholm Förh.*, **7**, 121.
- NOVÁČEK, R. (1935), Study on some secondary uranium minerals: *České Spol. Nauk. Věstník II* (1935).
- PAGE, L. R. (1950), Uranium in pegmatites: *Econ. Geology*, **45**, 12.
- PALACHE, C., AND BERMAN, H. (1933), Oxidation products of pitchblende from Great Bear Lake: *Am. Mineral.*, **18**, 20.

- QUENSEL, P. (1940), Minerals of the Varuträsk pegmatite XIX. The uraninite minerals: *Geol. fören. Stockholm Förh.*, **62** 391.
- RAGSKY, F. (1853), *Annalen der Physik*, **4**, 348.
- RIGAL, R. (1938), Las minas de columbita en la Cañada de Alvarez: *Argentina Bol. Dirección de Minas y Geología*, no. **45**.
- ROBERT, M. (1940), The geology of the Katanga: *Inst. royal colonial belge, Mem.* **6**, Fasc. **2**, 1-108.
- ROSS, C. S., HENDERSON, E. P., AND POSNJAK, E. (1931), Clarkeite, a new uranium mineral: *Am. Mineral.*, **16**, 213.
- SARKAR, P. B., AND SEN, R. K. (1946), Application of x-ray diffraction methods in the analysis of an ore from Rajputana: *Science and Culture*, **11**, 388.
- SARKAR, T. C. (1941), The alteration of radioactive minerals: *India Acad. Sci., Proc.*, **14B**, 261.
- SCHAEFER, T. (1847), Untersuchung einiger Mineralien, welche tantal säure-ähnliche Metallsäuren enthalten: *Ann. Phys.*, **72**, 570.
- SCHOEP, A. (1930), Les minéraux du gîte uranifère du Katanga: *Ann. Mus. Congo Belge*, ser. 1, pt. 2, 1.
- SCHOEP, A., AND DE LEENHEER, L. (1937), Qu'est-ce que la gummite?: *Soc. géol. belge Bull., Pal., Hydrol.*, **46**, 309.
- SCHOEP, A. (1921), Sur la présence du radium au Katanga: *Soc. chim. Belgique Bull.*, **30**, 219.
- SCHOEP, A. (1936), Sur les associations à cuprosklodowskite du Katanga: *Soc. géol. belge Bull.*, **46**, 298.
- SHAUB, B. M. (1938), The occurrence, crystal habit and composition of uraninite from the Ruggles mine, Grafton Center, N. H.: *Am. Mineral.*, **23**, 334.
- STEINKUHLER, W. (1923), Sur pechblende du Congo Belge: *Soc. chim. Belgique Bull.*, **32**, 233.
- STEINOCHEK, V., AND NOVÁČEK, R. (1939), On beta-uranotil: *Am. Mineral.*, **24**, 324.
- STRUNZ, H. (1949), Mineralogische Tabellen, 2d ed., Leipzig.
- TIPPER, G. H. (1919), On pitchblende, monazite and other minerals: *Indiana Geol. Survey Rec.*, **50**, 256.
- THOREAU, J., AND DE TERDONCK, R. DU T., (1933), Le gîte d'uranium de Shinkolobwe, Katanga: *Inst. colonial Belgique, Mém.* **1**, fas. 8 (1933); *Inst. royal belge, Sect. Sci. Nat., Mém.* **2**, ser. 4, 3 (1933).
- THOREAU, J., AND DE TERDONCK, R. DE T. (1935), Concentrations uranifères du Katanga: *16th Internat. Geol. Congress.*
- VERNADSKY, W., AND CHAMIE, C. (1924), Sur une pseudomorphose de la curite: *Comptes Rendus*, **178**, 1726.
- WASSERSTEIN, B. (1951), Cube-edges of uraninite as a criterion of age: *Nature*, **168**, 380.
- WASSERSTEIN, B. (1954), The ages of uraninite by a new method: *Nature*, **174**, 1004-1005.
- WEBSKY, C. F. M. (1853), Ueber die geognostischen Verhältnisse Kupferberg: *Deutsche geol. Gesell. Zeits.*, **5**, 427.
- WEBSKY, C. F. M. (1859), Ueber Uranophane: *Deutsche geol. Gesell. Zeits.*, **11**, 384.
- WHITNEY, J. D. (1849), Chemical examination of some minerals, coracite: *Am. Jour. Sci.*, ser. 2, **7**, 434.
- YAGODA, H. (1946), Radiocolloid aggregates in uranium minerals: *Am. Mineral.*, **31**, 462.

Manuscript received Nov. 25, 1955.