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RADIOACTIVE COLUMBITE[1]

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Abstract

Columbites may be radioactive because: 1) they contain isomorphous U^{4+}(and Th^{4+}?) substituting for Fe^{2+}; 2) they contain growth inclusions of other radioactive species (*e.g.*, monazite); 3) they contain exsolved uraninite; 4) they are overgrown or partly replaced by other hypogene radioactive species (*e.g.*, samarskite); 5) they are coated or veined by secondary uranyl minerals (*e.g.*, parsonsite); 6) of various combinations of 1–5. Columbite from British Columbia (9–10% U_3O_8), although apparently homogeneous when examined by means of *x*-ray diffraction methods, is shown to be a mixture of non-radioactive columbite and radioactive, metamict pyrochlore by means of an electron microprobe analysis. The material toddite is shown to be a mixture of columbite and samarskite.

Radioactive columbite is related to a particular pegmatite district, its paragenetic position, and the presence of other accompanying radioactive species.

Introduction

In the early 1950's the writer began a general investigation of the properties and textures of the opaque pegmatite minerals in polished section. Species studied included columbite-tantalite, stibiotantalite, samarskite, euxenite, allanite, uraninite, fergusonite, gadolinite, magnetite, ilmenite and various sulfides. In the course of the study it was found that several of the columbites were radioactive to varying degrees. This satellitic problem was enhanced in 1955 by the discovery, at the Mica Lode pegmatite in Fremont County, Colorado (Heinrich, 1948), of a relatively large body of columbite-bearing rock. From this ore body 1,200 lbs of columbite was eventually recovered, which reportedly assayed 3.2% U_3O_8.

The preliminary results of the study were published as an abstract (Heinrich and Giardini, 1956). Since 1956 several other investigators have contributed both scattered observations and systematic data, and this paper not only presents my own results but also correlates and summarizes the work of others on radioactive columbite.

The writer is indebted to Dr. A. A. Giardini and to S. H. Quon for assistance in the laboratory and to Drs. S. C. Robinson, Clifford Frondel and V. B. Meen for the loan and gift of specimens. Dr. Wm. C. Kelly also kindly prepared several samples by means of the microdrill and identified them. Both the field and laboratory studies were supported by grants from The University of Michigan, Phoenix Memorial Project, Nos. 150 and 204.

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PREVIOUS STUDIES

As nearly as can be determined the first record of uranium in columbite is in the results of the analysis presented by Genth (1889) of a columbite from Mineral Hill, Pennsylvania. Toddite was described by Ellsworth (1926) as a uraniferous variety of columbite, but results presented in this paper do not support this designation. Hutton (1959) has established the existence of metamict columbite. Data on the previously reported occurrences of radioactive columbites are summarized in Table 1.

RESULTS OF INVESTIGATIONS

General

All of the columbites described were studied in polished sections prepared by the means of a Graton-Vanderwilt lap. Identities, where in doubt from optical studies, were checked by means of *x*-ray powder data. Specific gravity determinations were made on the Berman microbalance, and most of the results represent the average of nine determinations made on three different fragments. Autoradiographs were prepared from the polished sections, first on alpha-particle plates and later on *x*-ray film. Most of the specimens were collected by the writer.

Eight-Mile Park, Fremont Co., Colorado
Ref.: Heinrich, 1958
Deposits represented: Mica Lode, Meyers Quarry

Columbite occurs as tabular crystals in a unit of albite-muscovite-beryl (± triplite and spessaritite) rock that replaces footwall parts of quartz-microcline cores. G (Mica Lode) 5.58, 5.66, and in the same specimen 5.69 and 5.74, indicating some Nb/Ta variation from specimen to specimen and also within individual crystals. G (Meyers Quarry) 5.67. All specimens weakly to strongly radioactive. Radioactivity highly variable. The patterns indicate most of the radioactivity stems from coating and fracture fillings of a secondary uranium mineral (Fig. 1a). Some specimens have the coatings but also contain primary disseminated inclusions of uraninite (Fig. 2a). In p.s. these tabular inclusions show local irregular variations in reflectivity (variation in U^{4+}/U^{6+} ratio), and some contain minute hair-like wisps of galena. Other specimens are homogeneous in p.s. or show narrow darker borders along fractures (oxidation of Fe^{2+} and Mn^{2+}?).

Thin sections of the Mica Lode columbite rock reveal the coatings and fracture fillings consist of a high-relief, high birefringent yellowish secondary uranium mineral in exceedingly fine-grained aggregates. *X*-ray powder diffraction data indicate that this mineral is parsonsite [strongest lines at d 4.24 (10), 3.29 (8), 2.14 (5), 3.95 (4), 3.45 (4), 1.73 (4)].

Devils Hole, Fremont Co., Colorado
Ref.: Hanley *et al.*, 1950

Thinly tabular columbite occurs in albitized parts of the intermediate zone. G 5.47, 5.52. No radioactivity detected. Homogeneous in p.s. except for a few veinlets with hematite plates.

Phantom Canyon, Fremont Co., Colorado
Ref.: Hanley *et al.*, 1950

Small plates of columbite occur with beryl in wall zone pegmatite. G 6.26. Radio-

TABLE 1. PREVIOUSLY REPORTED RADIOACTIVE COLUMBITES

Locality	Radioactivity	Deposit	Reference
Mineral Hill, Penn.	0.18% UO_3	pegmatite	Genth, 1889
Woodcox Mine, Hastings Co., Ontario, Canada	3.44% ThO_2	pegmatite	Ellsworth, 1932
Verity deposit, Blue River Region, B. C., Canada	9–10% U_3O_8	carbonatite	Robinson, writt. comm. 1955, 1956; Rowe, 1958
Hagendorf, Bavaria, Germany	0.15% U	pegmatite	Scholz, 1925, Zierh, 1957, Ramdohr, 1958
Hühnerkobel, Bavaria, Germany	$<0.01\%$ UO_2 autoradiograph gave pale uniform darkening in 30 days	pegmatite	Strunz and Tennyson, 1961
Ilmen Mtns., Urals, USSR	0.54, 0.56, 0.50% U_3O_8	pegmatite	Kuznetsov, 1956
Congo (Belgian)	0.01–0.1% U (11 anal.) aver. 0.5%	pegmatite	Van Wambeke, 1955
Northern Lugulu, Kivu, Congo (Belgian)	0.03–0.41% UO_3	pegmatite	de Kun, 1959
Umeis, Warmbad Dist., South-West Africa		biotite schist flanking pegmatites	Pike, 1958
Baragoi Dist., Kenya		pegmatite	Haverson and Pulfrey, 1953
Matlabas River, Waterberg Dist., Transvaal		carbonatite	Pike, 1958
Ambatofotsikely, Malagasian Republic	2.02% U_3O_8	pegmatite	Turner, 1928
Odara, Central Travancore, India	0.25% eU 0.25% U_3O_8 0.29% U_3O_8	pegmatite	Paulose, 1950, 1957; Paulos and Mathai, 1951
Gaya Dist., India	0.81% U_3O_8 1.7% U_3O_8	pegmatite	Nag, 1929
Tan-yo-men and Sekisyo-men, Korea	tr-0.05% UO_2	pegmatite	Hata and Iimori, 1938
Yinnutharra, Australia	0.96% UO_2, metamict 0.86% U 0.059% ThO_2	pegmatite	Hutton, 1957, 1959

activity generally nil except for a very few small irregular (primary?) inclusions. In p.s. a few darker inclusions and small irregular areas of higher reflectivity (higher Ta content?).

Meyers Ranch, Park Co., Colorado
Ref.: Hanley *et al.*, 1950

Columbite forms thick tablets with muscovite and beryl in an albitized core-margin replacement unit. G 5.62, 5.79. Radioactivity very weak. A very slight *uniform* darkening

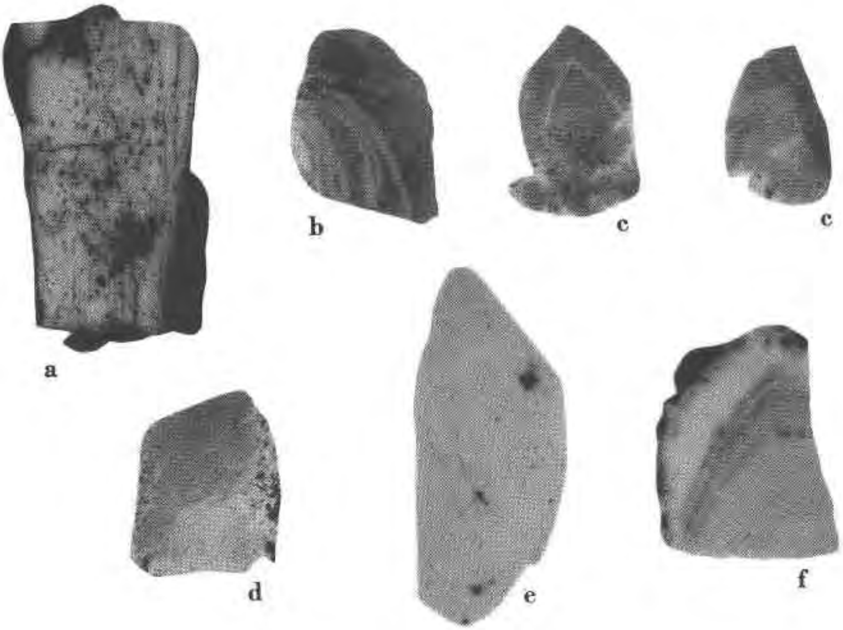

FIG. 1—Autoradiographs of radioactive columbites. (×1) a) Mica Lode, Eight Mile Park, Fremont Co., Colo. Coatings and veinlets of parsonsite; exsolution (?) blebs of uraninite. b) Apache, Petaca, N.M. Semi-regular variation in radioactivity. U isomorphously held. c) Cribbenville, Petaca, N.M. Isomorphous U variable in crystallographic zones. d) Eureka, Petaca, N.M. Zonal variations in isomorphous U (core and broad outer zone) and marginal inclusions and minute veinlets of samarskite (black). e) McKinney, Spruce Pine, N. C. Inclusion and very thin veinlets of samarskite. f) Prince Edward Co., Va. Crystallographically zoned U and replacement overgrowth and veinlet of euxenite.

produced after 44 days exposure. Uniformly reflective and inclusion-free or with widely scattered minute, irregular blebs of hematite in p.s.

Brown Derby Area, Gunnison Co., Colorado
Ref.: Hanley *et al.*, 1950, Straatz and Trites, 1955
Deposists represented: Brown Derby No. 1, Brown Derby No. 4, Nesbit, White Spar No. 1.

Columbite is in thin to thick tablets usually in wall zone rock. In the B. D. No. 5 it is associated with monazite, gahnite and zinnwaldite. G 5.61 (B.D. No. 4), 5.63 (B.D. No. 1), 5.70 (Nesbit), 5.85 (W.S. No. 1). No radioactivity detected after 30 day exposures. Generally homogeneous in p.s. except for 1) darker "oxidation" veinlets and outer shells and 2) irregular to rounded scattered inclusions of cassiterite (Fig. 2b).

Burroughs pegmatite, Kitteridge, Jefferson Co., Colorado
Ref.: Hanley *et al.*, 1950

Thickly tabular columbite (G 5.70) is relatively homogeneous in p.s. Near the margins appear irregular to elongate areas of distinctly higher reflectivity (higher Ta content?). Radioactivity very weak; a 44-day exposure produced a very faint *uniform* darkening.

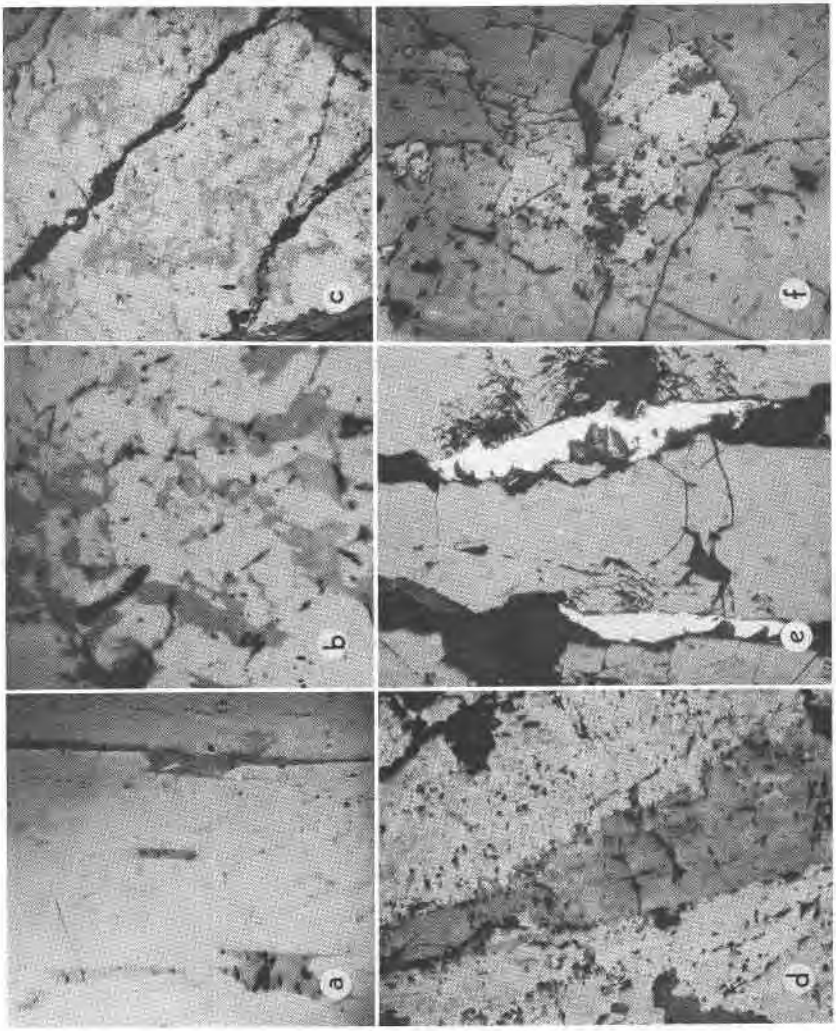

FIG. 2—Photomicrographs of radioactive columbites, reflected light. (a, b, c, d, f—×80; e—×160) a) (Upper row, left) Mica Lode, Eight Mile Park, Fremont Co., Colo. Exsolution(?) tablets of uraninite. b) (Upper row, center) Brown Derby No. 1, Gunnison Co., Colo. Irregular inclusions of cassiterite and a non-metallic species (black). c) Eureka, Petaca, N.M. Marginal irregular replacement of columbite (lightest gray) by metamict samarskite (variable medium gray). Late veinlets of quartz (black). c) Tin Mtn., Black Hills, S. D. Columbite and tapiolite minutely intergrown (both sides of central darker oxidized band). e) Newry, Me. Veinlets of galena. f) Lyndoch, Renfrew Co., Ont. Remnants of columbite (lighter gray) in metamict euxenite (more abundant, darker, variable gray).

Whippet No. 3, Fremont Co., Wyoming
Ref.: Hanley *et al.*, 1950

Tantalite occurs as thick stubby crystals in a lepidolite-rich replacement unit. G 6.21, 6.56. No detectable radioactivity. In p.s. there appears a narrow marginal zone of distinctly lower reflectivity (very likely an oxidation shell).

Placerville, Idaho

Columbite occurs as subangular placer pebbles. G 5.34. Radioactivity not detectable. Homogeneous in p.s. except for numerous gangue inclusions, chiefly quartz. Deep red-brown internal reflection indicates high Mn content.

Petaca District, New Mexico
Ref.: Jahns, 1946
Deposits represented: Apache, Cribbenville, Eureka, Globe, Hidden Treasure, Lonesome, North Star, Werner

Columbite occurs chiefly in strongly albitized parts of the pegmatites.

Apache: G 5.60. Strongly and variably radioactive. Some specimens show moderate general darkening with curving strips of somewhat lower radioactivity (Fig. 1b). Another displays general darkening with irregular patches and veinlets with lesser radioactivity and small irregular inclusions of distinctly higher radioactivity. Considerable variation in reflectivity in p.s., with a darker phase occurring as irregular veinlets and patches. The darker phase, which is the more radioactive, apparently represents columbite of lower Nb content. A mixture of 50% HF+50% conc. H_2SO_4 applied for two minutes etches the darker phase very strongly, leaving the lighter phase essentially unaltered.

Cribbenville: G 5.52. Moderately radioactive. Strong variations in radioactive in crystallographically oriented zones (Fig. 1c). In p.s. relatively homogeneous, with a few small darker patches and veinlets unrelated to the growth zones and a few marginal inclusions of cassiterite.

Eureka: G 6.00. Moderately radioactive with zonal variations (Fig. 1d) and a narrow outer zone rich in highly radioactive particles. Homogeneous in polished section except for the margins which are partly replaced by samarskite which extends inward locally from the margins of the columbite crystal as an irregular network and also along fractures (Fig. 2c). The samarskite is metamict.

Globe: "Feather" and blocky types (G 5.82). The blocky crystals show weak radioactivity of uniform distribution. The "feather" type (Fig. 3) has no detectable radioactivity. Both are homogeneous in p.s.

Hidden Treasure: G 5.58. Very weak uniform radioactivity. Homogeneous in p.s.

Lonesome: G 5.96, 5.99 in the same specimen. Strongly radioactive.

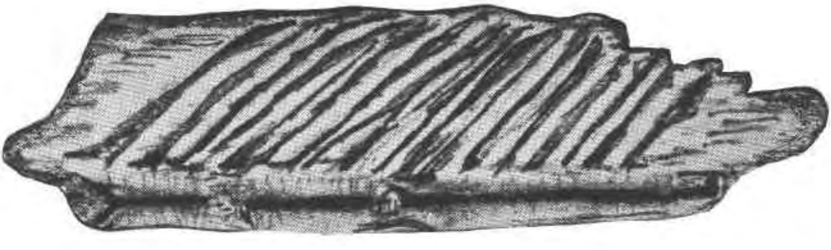

FIG. 3—"Feather" columbite, Globe mine, Petaca, N.M. (×2).

North Star: G 5.19, 5.26 in same specimen. Moderately radioactive and uniformly so, except that in some crystals there are concentrations of minute highly radioactive inclusions marginally or along fractures. In p.s. these inclusions show lower reflectivity than the columbite, variable reflectivity, and strong variable reddish internal reflection—samarskite. Otherwise the columbite is homogeneous.

Werner: G 5.41 Moderately and uniformly radioactive. Homogeneous in p.s.

Harding Mine, Taos Co., New Mexico
Ref.: Montgomery, 1950

G 6.19 (Specimens studied), 6.55 (composite, ore mined). No detectable radioactivity· Generally homogeneous in p.s. but locally shows small highly irregular areas of darker gray color, with brownish internal reflection (probably higher Nb and Mn contents), unrelated to fractures or margins.

Black Hills, South Dakota
Ref.: Page *et al.*, 1953
Deposits represented: Bob Ingersoll, Beecher, Old Mike, Tin Mtn., "Keystone" (2), "Pennington Co."

Bob Ingersoll: G 6.1, 6.5 in same specimen. No detectable radioactivity. Completely homogeneous in polished section save for gangue inclusions.

Beecher: G 5.46, 5.53 in same specimen. Very weakly and uniformly radioactive except along veinlets which contain gangue minerals and goethite. In p.s. homogeneous except for the goethite veinlets.

Old Mike: G 7.72. No detectable radioactivity. Homogeneous in p.s. except for very minor irregular blebs of slight darker color (lower Ta?).

Tin Mtn.: G 6.14–6.79 in same specimen. No detectable radioactivity. In p.s. seen to consist of about 80% columbite and 20% intergrown tapiolite in crystals and tiny blebs (Fig. 2d). Some ore sold had 55% Ta_2O_5; another lot had 75% Ta_2O_5. *X*-ray methods (Page *et al.*, 1953, p. 173) confirmed tapiolite in specimens with G 7.32, 7.64.

Three other Black Hills specimens all showed no radioactivity and except for gangue inclusions were homogeneous in p.s.

Spruce Pine District, North Carolina
Ref.: Olson, 1944
Deposits represented: McKinney, Deer Park No. 5

G 5.36, 5.64. All specimens very weakly radioactive. In some McKinney columbite are small included crystals of highly radioactive samarskite (Fig. 1e) around which, in p.s., alteration haloes, similar to those described by Ramdohr (1958) can be seen. Also a few thin "oxidation" veinlets. Otherwise homogeneous in p.s.

Prince Edward Co., Virginia

Through the courtesy of Prof. R. S. Mitchell, I was able to study three specimens of radioactive columbite from east of Darlington Heights. The autoradiograph (Fig. 1f) shows that the radioactivity stems from two sources: 1) isomorphously contained uranium which is distributed variably in zones and 2) highly radioactive irregular border and minor fracture fillings. In p.s. the central part of the columbite crystal appears essentially homogeneous, but the radioactive border can be seen to consist of two phases: irregular, small remnants of columbite in a replacing mineral that shows highly variable but generally low reflectivity; strong, minute fracturing and strong variable yellow-brown internal reflection. It is metamict and most likely is euxenite.

Haddam, Connecticut
Ref.: Cameron *et al.*, 1954

G 6.21. No detectable radioactivity. Homogeneous in p.s.

Newry, Maine
Ref.: Cameron *et al.*, 1954

G. 5.70. No detectable radioactivity. Homogeneous, except for small amounts of galena along some fractures (Fig. 2e).

Lyndoch, Renfrew Co., Ontario
Ref.: Ellsworth, 1932

$Ta_2O_5 = 55.79\%$. Specimens are thinly bladed. In p.s. they are seen to be a composite of columbite partly replaced by euxenite ("lyndochite") (Fig. 2f). The columbite parts are not radioactive, whereas the euxenite is strongly so. It is doubtful whether the analysis represents the true Ta content of the columbite. In specimens of "lyndochite" Ellsworth (1932) reports columbite remnants.

Rabenstein, Bavaria, Germany

No detectable radioactivity. In p.s. homogeneous.

INHOMOGENEITY IN COLUMBITE-TANTALITE

That the Nb/Ta ratio of columbite-tantalite decreases with relative decreasing age in paragenetically distinct series members from individual pegmatites has been established by numerous investigators (*e.g.* Björlykke, 1935, 1937; Quensel and Berggren, 1938; Hanley, 1947; Cameron *et al.*, 1949; Heinrich, 1953; Ginzburg, 1956). Landes (1925) appears to have been one of the first to present data leading to this conclusion. In many instances this decrease in Nb/Ta is accompanied by a decrease in Fe/Mn (Heinrich, 1953, Ginzburg, 1956). Furthermore the general sequence of Nb/Ta species in pegmatites also has been clearly determined as (Heinrich, 1953; Ginzburg, 1956):

1. (ferroan) columbite
↓ (tapiolite)
2. (manganoan) tantalite
3. thoreaulite
4. stibiotantatite, bismutotantalite
5. simpsonite
6. microlite (behierite?)

Examples are known of stibiotantalite replacing tantalite (Greenbushes, Australia), microlite replacing tapiolite (Strelley, Australia), microlite overgrown on tapiolite (Kimito, Finland), as well as other textural and geological relationships in support of this sequence.

Less attention has been given to the homogeneity of single grains or crystals of columbite-tantalite. It has generally been assumed that a specific gravity determination is an accurate measure of the Nb/Ta ratio, although serious discrepancies have appeared. (In some of the specimens described above, *e.g.* Mica Lode, Lonesome, North Star, Bob Ingersoll, Beecher, Tin Mtn., the specific gravity varies appreciably in different parts of single crystals.) Charts embodying this relationship have been prepared by Mügge (1924), Pehrman (1932), Simpson (1936), Slavin *et al.* (1946), Jahns (1946), and Campbell and Parker (1955).

Causes of inhomogeneity in single crystals of columbite-tantalite are:

1. Variations in Nb/Ta ratio
   a. Zonal growth
   b. Irregular (replacement)
   c. Exsolution?
2. Inclusions
   a. Non-metallic species. "Gangue"-quartz, plagioclase, microcline, etc. (Growth inclusions and replacement).
   b. Metallic and semi-metallic species
      1) Exsolution
      2) Growth (contemporaneous)
      3) Replacement
3. Alteration, $Fe^{2+} \rightarrow Fe^{3+}$, $Mn^{2+} \rightarrow Mn^{3+}$
   a. Weathering
   b. Hydrothermal (?)
   c. Radioactive haloes

Isküل and Kurbatov (1935) described crystals with zonal and exsolution (?) structures and pointed out that the reflectivity (in p.s.) in the series increases with increasing Ta content. The exsolution structures were described as generally characterizing marginal parts of crystals but locally appearing in interior parts or even in veinlets. Some of these may well be replacement features like those described by Ginzburg (1956) who demonstrated that many columbite-tantalites have zonal structure and that many others are complexly and irregularly heterogeneous owing to replacement of columbite by columbite-tantalite and/or manganoan tantalite. Primary (?) intergrowths with tapiolite also are known (Tin Mtn.).

In addition, columbite-tantalite or tantalite may be replaced by microlite, stibiotantalite, tapiolite, samarskite (Petaca district, N. M.) and euxenite (Lyndoch, Ont., Virginia). Metallic species recorded as inclusions in columbite-tantalite are: cassiterite, galena, hematite, ilmenite, rutile, samarskite, tapiolite, uraninite. Of these, cassiterite, ilmenite, rutile and uraninite may have resulted from exsolution in some cases.

There is little information on the oxidation of columbite-tantalite, but some specimens in p.s. show thin, darker, irregular zones which extend inward along fractures and may form borders along gangue veinlets. This lower reflecting material apparently represents columbite-tantalite in which Mn^{2+} and Fe^{2+} have been oxidized. It is clearly secondary, and its formation is initiated at grain surfaces or boundaries. It is etched more readily by 50% HF+50% conc. H_2SO_4 than is ordinary columbite. It is possible that in some cases such alteration may be the result of the action of late-stage hydrothermal solutions rather than that of supergene waters. Ramdohr (1958) has described haloes of alteration around inclusions of uraninite in columbite, and these occur in McKinney columbite around samarskite. Presumably these haloes also are characterized by this oxidation result. Goethite veinlets are known to cut columbite.

RADIOACTIVITY IN COLUMBITE

On the basis of the results obtained from autoradiographs and polished sections and from previously reported observations, radioactive columbite-tantalites are of the following genetic types:

I. Uranium and/or thorium within the crystal structure
  A. Uniformly distributed
  B. Variable, in zones
II. Inclusions of other hypogene radioactive species
  A. Syngenetic ("growth") inclusions:
    1. Uraninite
    2. Cyrtolite
    3. Monazite
  B. Exsolution inclusions
    1. Uraninite
III. Replacement or overgrowth by hypogene radioactive species:
    1. Uraninite
    2. Samarskite
    3. Euxenite
    4. Microlite
IV. Coating and veining by secondary radioactive species:
    1. Parsonsite
V. Various combinations of I–IV.

This classification is an elaboration of that presented briefly by Heinrich and Giardini (1956). Van Wambeke (1956) recognized three types of radioactive columbite occurring in Congo pegmatites:

1. With U in the crystal structure.
2. With secondary yellow uranium minerals as coatings and along cleavages. The U could have been derived from the columbite.
3. With inclusions of uraniferous (Ti)-Nb-Ta minerals.

Isomorphous U and Th

Examples of columbite-tantalites with isomorphous U uniformly distributed include those from: Hühnerkobel, Bavaria; Yinnutharra, Australia (manganomossite); Burroughs pegmatite, Colorado; the Globe, Hidden Treasure, Lonesome, North Star, and Werner deposits, Petaca District, New Mexico; and the Beecher Lode, South Dakota. Examples of deposits containing columbite-tantalite with zonal variations in radioactivity are: Congo; and unknown Australian locality (manganomossite); the Cribbenville and Eureka pegmatites, Petaca District, New Mexico; and Prince Edward Co., Virginia. The amount of uranium that can be retained in the structure varies considerably, from 0.01–1% U_3O_8. Columbites with higher U contents are reported (Table 1), but their homogeneity has not been checked. Van Wambeke (1955) found that Congo columbites rarely contain more than 0.6% U. The radioactivity of specimens that produced *some* autoradiographic darkening examined by the writer ranged from nil to 0.7% eU_3O_8.

Despite such designations in the literature, it seems unlikely that *uranian* columbites exist. The presence of primary inclusions of uranite and of exsolved(?) uraninite blebs indicate that the uranium exists as U^{4+} rather than U^{6+}. Thus these uraniferous columbites are *uranoan* columbites. The substitution in the structure may be one of the following types:

$Fe^{2+}Nb^{5+} \equiv Nb^{5+}U^{4+}Ti^{4+}Ti^{4+}$
$Fe^{2+}Nb^{5+}Ta^{5+} \equiv U^{4+}Ti^{4+}Sn^{4+}$
$Fe^{2+}Nb^{5+} \equiv U^{4+}Al^{3+}$
$Fe^{2+}Nb^{5+} \equiv U^{4+}Fe^{3+}$

It seems unlikely that U substitutes for Nb, despite the statement by Strunz and Tennyson (1960) that " . . . das hierfür verantwortlich Uran wird dadoch für Nb oder Ta in statistischer Verteilung in Columbitgitter anwesend sein."

Whether thorium can also be a significant isomorphous element in columbite-tantalite is uncertain. In Congo material thorium was not detected, with a sensitivity of a little less than 0.01% Th (Van Wambeke, 1955). Hutton (1959) reports the Australian "manganomossite" contains 0.059% ThO_2. However, Ellsworth (1932) cites a columbite from the Woodcox Mine, Ontario, that contains 3.44% ThO_2. This material requires re-examination for homogeneity.

That radioactive columbite can undergo structural damage to the extent of becoming nearly completely metamict is demonstrated by the Hutton's study (1959) of "manganomossite," which he found to be metamict with but 0.96% UO_2. Recrystallization upon ignition restored the columbite structure. It remained to be explained why the material from British Columbia with 9–10% U_3O_8 had not become metamict. A particularly careful examination of the x-ray powder film for this columbite showed no uraninite lines and, in fact, no lines that could not be assigned to columbite. Thus, the homogeneity of the material seemed certain. Unfortunately the particles were too small for p.s. study. However, a nagging doubt as to the homogeneity of the sample lingered; and an electron microprobe analysis was performed on the minute grains. The electron micrograph shows that the apparently homogeneous particles consist almost entirely of two distinctly different minerals, complexly intergrown. The more abundant species (A) is the lighter, the less abundant (B) the darker.

The probe reveals that A contains high concentrations of Nb and Fe; B has high concentrations of U, Ca and Ti as well as appreciable amounts of Nb. Species A is the columbite from the composition and *x*-ray data. It is concluded that species B is completely metamict uranoan pyrochlore. Three things support this identification: 1) the composition, 2) the absence of *any* pyrochlore lines in the *x*-ray powder photograph, and 3) uranoan pyrochlore also occurs in the Verity deposit as disseminated crystals containing 6% U_3O_8 (Rowe, 1958, p. 33).

A Si scan revealed a few inclusions rich in this element, probably quartz.

The results of the microprobe study thus are:

1. The Verity uraniferous columbite is a submicroscopic mixture of non-radioactive columbite and uranoan pyrochlore. This is why this columbite has not become metamict whereas the Australian "magnanomossite" with only about 1% UO_2 is metamict.
2. *X*-ray diffraction results cannot differentiate between a completely homogeneous sample and a mixture, *if the mixture consists of a crystalline phase and a metamict phase.*

Radioactive inclusions

Primary (syngenetic) inclusions of radioactive minerals are represented by the species, uraninite (Hagendorf, Bavaria; Mica Lode, Colorado); and cyrtolite and monazite (Ginzburg, 1956). The uraninite in the Hagendorf columbite forms cubic crystals up to 0.05 mm and octahedra up to several mm (Ziehr, 1957). Doubtless other radioactive species can also serve in this capacity, and probably some other radioactive columbites not studied autoradiographically belong in this category. Ramdohr (1958) considers the fine-grained uraninite not near the crystal margins in the Hagendorf columbite to have resulted from exsolution. This may also be the case for the Mica Lode material.

Radioactive replacements or overgrowths

Overgrowths of uraninite on columbite are known from Hagendorf (Strunz, 1961). Replacements of columbite are represented 1) by samarskite in the Eureka and North Star material, Petaca, New Mexico and the McKinney material, North Carolina; 2) by euxenite in the Lyndoch, Ontario and Virginia specimens; and 3) by pyrochlore-microlite, which also may be distinctly radioactive and is known to replace columbite in several districts in the world.

Secondary radioactive species

The parsonsite coatings and cleavage and fracture fillings on the Mica Lode columbite exemplify this type of material. Whether the parsonsite is supergene in this deposit is uncertain. The parsonsite-bearing columbite occurs with quartz, albite, muscovite, oxidized spessartite, partly kaolinized beryl and altered triplite.

PARAGENETIC SIGNIFICANCE

The presence and amount of isomorphous uranium in columbite appears to be a feature characteristic for a particular pegmatite district (Table 2). Whether or not a columbite will be radioactive may also be a function of its paragenetic position. Ginzburg (1956, p. 315) states:

> "As a rule, crystals of thick tabular columbite are more or less radioactive. Their radioactivity is caused by minute inclusions of uraninite . . . or inclusions of cyrtolite, or rarer, inclusions of monazite."

This type of columbite, which is Ta-poor, occurs in muscovite-beryl pegmatite and is regarded as of magmatic, rather than hydrothermal origin. Younger manganoan tanalite may be radioactive owing to inclusions of cyrtolite or microlite.

The Apache material (Fig. 1b) is instructive. The darker, less reflective phase, which is presumably of lower Nb content, is the more radioactive. The Nb content of columbite decreases with decreasing relative age and thus with decreasing temperature of formation. It is to be expected, that with decreasing temperature of crystallization, lesser amounts of uranium could be accommodated in the columbite-tantalite structure. Therefore there may be a *general* variation of isomorphous U with the Nb/Ta ratio.

The presence of other uranium-bearing minerals in the deposit also

TABLE 2. DISTRICT VARIATION OF RADIOACTIVITY (ISOMORPHOUS U) IN COLUMBITE
S = strong, M = moderate, W = weak, BD = barely detectable

Radioactive	Non-radioactive
Eight Mile Park, Colo. (M)	Devils Hole, Colo.
Meyers Ranch, Colo. (W-BD)	Phantom Canyon, Colo.
Burroughs, Colo. (BD)	Brown Derby, Colo.
Petaca, N. M. (S-M)	Fremont Co., Wyo.
Spruce Pine, N. C. (W)	Harding, N. M.
Prince Edward Co., Va. (M)	Black Hills, S. D. (nil-BD)
	Haddam, Conn.
	Newry, Maine
	Lyndoch, Ont.

appears to increase markedly the possibility of uranium entering the structure of columbite.

Finally, it is to be noted that radioactive columbites have been found in three general types of deposits: 1) pegmatites, 2) exomorphic zones around pegmatites and 3) carbonatites.

TODDITE

Toddite was described by Ellsworth (1926) from a pegmatite in Dill Township, Sudbury, District, Ontario. He stated (p. 334)

> "From the analysis it is evident that the mineral conforms in a general way to the columbite formula and may be considered as columbite in which some manganese and iron is (sic) replaced by uranium. . . . The microscopic examinations revealed no evidence of a lack of homogeneity."

Palache *et al.* (1944, p. 785–786) state that

> "Although this material has been considered to be a uranian-columbite, the extremely low specific gravity, the absence of cleavage and the relatively large content of rare earths, Ca, Fe^3, and water render such an interpretation improbable. Evidence of admixture has been found in some specimens" (Berman, priv. comm., 1940), "and the material may be a mixture of columbite and some other mineral, possibly euxenite."

Two specimens of material called toddite were obtained: 1) Harvard Museum No. 103937 "Toddite, Ellsworth's second locality." Professor Clifford Frondel (writt. comm.) states "I have no information about the specimen other than that given on the label"; 2) Royal Ontario Museum No. M-15510. Attempts to obtain some of Ellsworth's type material from the Geological Survey of Canada were unsuccessful. Dr. S. C. Robinson states (writt. comm.) " . . . Mr. Steacey . . . was able to find material from the toddite locality in Dill Township but *x*-ray checks of all possible minerals in it failed to identify toddite."

Both the Harvard and Toronto specimens were somewhat less than minuscule in amount—just sufficient for *x*-ray powder work.[1] Both gave but a few diffuse lines when *x*-rayed without being heated, and they were certainly largely metamict. Upon heating at 1000° C. for 1 hour in air, fair patterns were obtained. The comparison presented in Table 3 shows that toddite is a mixture of columbite and samarskite, and thus the name, toddite is invalid both as a species and a varietal designation.

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[1] Frondel (writt. comm.) described the specimen, "The seemingly empty vial accompanying this letter contains our stock of toddite."

Table 3. Comparison of x-ray Powder Date for Toddite with Columbite and Samarskite. CuKα Radiation Camera Diam. = 11.6 cm

1		2		3		4	
dÅ	I	dÅ	I	dÅ	I	dÅ	I
4.03	m			4.09	vvw(b)		
3.91	vvw	3.85	ms			3.91	5
3.60	vvw			3.64	m		
		3.48	m			3.48	5
3.22	m	3.20	m				
3.15	vvw					3.16	7
3.08	vvw	3.10	w				
3.00	s	2.95	m	2.96	s	2.94	9
2.91	vw	2.85	m	2.86	vw	2.84	9
2.79	vvw					2.74	2
2.67	vvw						
2.59	m	2.57	vw	2.54	vw	2.55	2
2.51	mw	2.49	ms	2.49	w	2.48	7
		2.44	w				
		2.37	w	2.38	vw	2.40	2
		2.30	vw	2.24	vvw	2.30	2
		2.10	w	2.09	w	2.15	5
2.01	vvw(b)	1.99	w			2.00	5
1.92	vvw(b)	1.90	w	1.90	vw	1.90	
1.86	vvw(b)					1.85	7
1.835	m	1.82	w	1.83	vw	1.82	
		1.76	w	1.77	mw		
		1.72	w	1.73	mw	1.72	5
1.70	w	1.699	w	1.72	m	1.69	7
1.69	vw					1.67	7
1.64	vvw					1.64	5
1.60	vvw(b)					1.62	5
1.58	vvw						
1.57	ms			1.54	mw(b)	1.56	10
1.50	w	1.48	vw	1.48	vw(b)	1.51	7
1.45	vvw			1.46	m(b)	1.49	7
1.43	vvw(b)	1.42	w			1.45	5
(15 others)							

1. Toddite, Royal Ontario Museum No. M-15510.
2. Toddite, Harvard No. 103937, "Ellsworth's second locality."
3. Columbite, Brown Derby No. 1 pegmatite, Gunnison Co., Colo.
4. Samarskite, USSR. ASTM.

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