

IDENTIFICATION OF METAMICT MINERALS BY X-RAY DIFFRACTION

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ABSTRACT

The physical properties of the metamict minerals are discussed and tabulated. Because of their isotropy and the variability of the index of refraction of these minerals, the petrographic microscope has limited value for their identification. Ordinarily, x -ray diffraction yields little additional information. Heating of certain specimens, however, induces some crystallinity in many of these "amorphous" minerals. A technique was developed for treating the metamict minerals to facilitate their systematic study for purposes of identification. Data are given for metamict zircon, thorite and uranothorite, euxenite, brannerite, fergusonite, samarskite, davidite, and allanite.*

It is also observed that some of the "more stable" related minerals that ordinarily occur in the crystalline state are structurally changed when subjected to temperatures at which many of the metamict minerals crystallize. This may indicate that the crystallized (by heating) metamict material is not reconstituted to the original crystal structure.

INTRODUCTION

Although the minerals that occur in the metamict condition have aroused increasing interest in recent years, little information has been published on methods of identification of the various minerals of this type. It is recognized that several naturally occurring substances having external crystallographic form nevertheless lack internal order and in many cases are amorphous to x -rays. The word "metamict" has been applied to these noncrystalline pseudomorphs of material presumed to have been crystalline originally. This term has been widely used, but it has been only recently that a detailed discussion of the subject has been published by Pabst (47). Brooker and Nuffield (10, 11) discuss the term as it has been applied to uraninite. These two readily available recent articles give a rather complete history and description of the word metamict, and nothing further on its origin and use will be included herein.

PHYSICAL PROPERTIES OF METAMICT MINERALS

The following list of physical properties of the metamict minerals is presented because it should be a useful guide to understanding of the difficulties and limitations of the problem of identification and classification.

* Tables are presented comparing in detail the x -ray diffraction patterns of these and related minerals after various heat treatments. An extensive bibliography of the general subject of metamictization is included.

1. *Optically isotropic and lacking in Debye-Scherrer X-ray diffraction* (2, 3, 4, others).

This is generally true. There are minerals, such as some allanites, however, that have a definite birefringence but give no x-ray diffraction pattern. Specimens are found in different stages of isotropism from relatively well crystallized to completely "amorphous"; some show heterogeneity in the same specimen. It is important to note that isotropism alone is no criterion as to the metamict condition of minerals that crystallize in the cubic system.

2. *Brittle with a conchoidal fracture and no cleavage.* Many of these non-crystalline "minerals" are clear, fresh appearing, and glassy. Some, however, appear altered and weathered in both the hand specimen and under the microscope. Many samples are filled with close fractures.

3. *Density increases when ignited.* This is usually true, but the difficulty of measurements on fractured or powdered material has probably led to some apparent contradictions. Consequently, this is not a particularly reliable criterion.

4. *Become incandescent on heating* (18, 36, 47, others). This phenomenon is included here, although it has been possible to crystallize all the metamict minerals studied by the author at temperatures appreciably below that required for glowing. In many instances this pyrognomic effect is not observed at any temperature.

5. *Crystallization by ignition below fusion or sintering temperature* (2, 6, 47, others). Crystallization may be very complex. With the possible exception of some zircons, no case of reconstitution of a single crystal has been observed. Some authors, however, have reported the re-formation of single crystals.

6. *Contain radioactive element* (20, 21, others). The radioactivity may be very low as in some allanites, gadolinites, and zircons; or it may be very high, as in thorites, brannerites, euxenites, etc. It is, however, apparently present in every instance in the metamict mineral. Not all radioactive minerals become metamict, as note thorianite, which is always found well crystallized.

This paper is not concerned with the causes and processes of metamictization, but only with the methods of identification of the metamict minerals. However, the author is firmly convinced that radiation from elements within predisposed minerals causes the metamict condition. Those references in the bibliography that are marked with an asterisk contain material on this phase of the metamict problem. The articles

by J. C. Slater (54) and F. Seitz (53) are particularly informative as to the effects of radiation.

METHODS OF IDENTIFICATION

Because of their isotropy and the variability of the index of refraction of metamict minerals the petrographic microscope has very limited value for their identification (17, others). Likewise, because of the complete disorder and consequent noncrystallinity of many of these naturally occurring compounds, *x*-ray diffraction yields little additional information (6, 47, 58, others). However, early in the study of these minerals mineralogists found that heating of certain specimens would give some crystallinity to many of these "amorphous" minerals (25, 46).

To make a systematic study of these heating effects, the following technique was developed (5).

1. All samples were handpicked with the aid of a microscope to remove contaminants.

2. Each sample was studied with the petrographic microscope to ascertain its general optical characteristics. Detailed optics were not determined because of the great variations occurring in similar specimens or sometimes in different parts of the same specimen.

3. Each sample was carefully split into several fractions to assure that the different tests would be run on identical materials.

4. One fraction was used for a semiquantitative spectrochemical analysis.

5. Other fractions were heat-treated as follows:

- (a) Crushed material was heated in an open crucible over a Meker burner for approximately 5 minutes.

- (b) Different portions of each specimen of crushed material were heated to different controlled temperatures in air for definite periods of time in an electric furnace.

- (c) Duplicate portions of the crushed material were heated to the same controlled temperatures in an inert atmosphere.

Difficulty was encountered in maintaining an inert atmosphere because it was found that both tank nitrogen and helium contained enough impurities to allow oxidation of the sample. Oxygen-bearing impurities were eliminated by passing the gas through a drying tube and then through a plug of heated steel wool before it reached the specimen being treated. This method had been suggested by F. A. Bannister and J. E. T. Horne (3).

DATA OBSERVED

It has been found that the only reliable method that can be used for the identification of the metamict minerals is the use of *x*-ray diffraction pat-

terns of heat-treated samples. Therefore, Tables 1 through 7, which contain pertinent and reproducible *x*-ray powder patterns of these minerals, are included. Where the author's results correspond to those previously published, the proper reference is noted and the data are not repeated here.

Since only semi-quantitative spectrochemical analyses were used as a check on the samples studied, the *x*-ray data may be open to some questions. However, the *x*-ray patterns tabulated herein are only those that have been reproduced on other samples from the same and other localities. No criterion was set up as to the number of different samples studied, but no table is included that has not been reproduced from at least five localities. For example, nearly identical fergusonite patterns have been produced by at least ten samples from different deposits.

The powder patterns of heat treated originally metamict zircon, thorite, and fergusonite can be indexed using the lattice parameters reported in the literature. However, attempts to index heat treated euxenite, davidite, brannerite, and samarskite were unsuccessful. Arnott (2) obtained similar (to those listed here) *x*-ray powder patterns of euxenite and states ". . . it was found that all but a few very weak lines could be satisfactorily indexed." Our data show some of these extraneous lines (which are invariably present) were too strong to be ignored. Efforts to relate these patterns to other ABO_4 or AB_2O_6 compounds (9) were likewise unsuccessful.

A. *Zircon*: (7, 15, 30, 35, 56, 61, others) (*x*-ray data in Table 1).

Specimens examined varied from completely "fresh," with sharp powder diffraction patterns in the small interplanar region as well as in the large interplanar region, through specimens with slight disorder (line broadening and poor diffraction in the small interplanar region), to those samples which appeared completely metamict and gave no powder diffraction pattern. Observations with the petrographic microscope disclosed that the fresh zircon had consistent optics that agreed well with those in the literature. In those specimens that are somewhat metamict, both the index of refraction and the birefringence decreased; complete isotropy was reached in the totally metamict specimens. Many zircons were studied, including the specimens labeled with the varietal names alvite, cyrtolite, and hagatalite. Heat treatment of these specimens generally resulted in well crystallized microcrystalline tetragonal zircon. However, there were exceptions and more detailed data on zircon have been included in another paper (35).

B. *Thorites and Uranothorites*: (8, 45, 46, others) (*x*-ray data in Table 2)

These minerals occur in nature in all stages of metamictization from

comparatively well crystallized to completely metamict material. X-ray diffraction data corroborated the petrographic data that all available thorites showed some degree of change to the metamict state.

The comparatively unchanged thorites give a powder pattern of the tetragonal zircon type. One of these samples studied has a unit cell that can be indexed with tetragonal lattice parameters of $a_0 = 7.15 \pm .01 \text{ \AA}$, $c_0 = 6.29 \pm .01 \text{ \AA}$. This material, heated to $1300^\circ \pm 20^\circ \text{ C.}$ in an inert atmosphere, gives an excellent pattern with $a_0 = 7.11 \pm .01$ and $c_0 = 6.32 \pm .01 \text{ \AA}$. The latter figures agree well with those reported by Pabst (46) for an unaltered uranothorite.

Pabst (45, 47) has discussed the dimorphism of thorium silicate and has presented data on the heat treatment of some thorites. The experiments carried out in our laboratories give data in general agreement with his, but some significant differences probably result mainly from variations in method of treatment of the original sample and possibly from the condition of the mineral before heat treatment.

These investigations disclosed the following results: when completely metamict thorites were heated to ca 800° C. for one minute or longer, face-centered cubic thorium oxide crystallized. The powder patterns of the heated samples always showed broad, diffuse lines resulting from either small particle size or internal disorder. When these same or similar samples were heated to higher temperatures (ca 1100° C.) in air for longer periods, tetragonal thorite or monoclinic huttonite was formed in addition to the thorium oxide. Spectrochemical analysis did not disclose any significant compositional difference in the specimens that crystallize differently when heat treated. Upon increase in temperature of heat treatment to ca 1350° C. in air, the original completely metamict material always became the monoclinic variety of thorium silicate plus some crystalline thorium oxide (present in all specimens ignited). When these samples were heated in a completely inert atmosphere at temperatures up to ca 1250° C. , the monoclinic dimorph of thorium silicate was formed in some and the tetragonal variety in others. However, if the temperature in the inert atmosphere is raised above ca 1350° C. , the tetragonal variety of thorium silicate plus some thorium oxide crystallizes and remains until the temperature of dissociation into thorium oxide and amorphous silica is reached at a temperature probably between 1600° C. and 1700° C.

Although the x-ray powder patterns of heated originally metamict thorium silicate differ in accordance with the method of heat treatment, our data indicate that this material can be definitely identified when heated under controlled conditions. However, evidence is as yet inconclusive as to whether the original mineral was huttonite or thorite.

C. *Euxenite*: (2, 14) (*x*-ray data in Table 3)

All euxenites examined and all that have been reported in the literature have been completely metamict. When powdered euxenites were heated in a crucible over a Meker burner, considerable variations in the resulting powder patterns occurred in material with almost identical composition. However, it was found that, when the same original material was heated at temperatures between ca 400° C. and 800° C. for extended periods (6+ hours), reproducible *x*-ray patterns could be made. Heating to ca 1200° C. in air for a short period (5+ minutes) produced sharp powder patterns that are likewise specific for euxenite but differ from those of the same samples heated at the lower temperatures. Ignition at this high temperature in an inert atmosphere produced still another structure. However, all three diffraction patterns are specific for the metamict mineral euxenite.

D. *Brannerite*: (48)

The brannerites examined were either completely metamict or produced only very weak and diffuse *x*-ray patterns that were useless for accurate identification. All metamict brannerites that were heated crystallized below ca 1000° C., and most of them became crystalline at temperatures somewhat less than 750°C. in only a few minutes. There is no discernible difference in the crystallization of the brannerites when they are ignited in an inert atmosphere.

E. *Fergusonite*: (4, 6) (*x*-ray data in Table 4)

Crystalline fergusonite has been reported from only one locality (60). This occurrence is of doubtful authenticity, and in all other described localities the mineral is found in the metamict state.

Fergusonite begins to crystallize at ca 400° C. and, if heated between this temperature and ca 800° C. for some time, will form a structure that can be indexed with a tetragonal unit cell ($a_0=5.18$, $c_0=5.48 \pm .02$ Å—Table 3). Because of originally poor diffraction patterns it was at first believed to have an orthorhombic cell (7). However, more precise measurements indicate that the tetragonal cell noted above is the correct one. If the temperature is raised above ca 800° C., fergusonite crystallizes with a structure identical to that formed when equimolecular proportions of Y_2O_3 and Nb_2O_5 are sintered or fused together. Barth (4) has indexed the pattern of this high-temperature modification on the basis of a tetragonal unit cell and states that it is the original fergusonite structure. The author believes, however, that more detailed work on both the high-temperature and low-temperature dimorphs described above is necessary

before a definite decision can be reached as to the original fergusonite structure.

It is of interest to note that if the a_0 of ignited fergusonite (ca 7.75 Å—Barth) is divided by the square root of 2 the result is nearly identical to what has here been indexed as the c_0 of this smaller tetragonal unit cell (5.48 Å). This suggests some structural relation between the two polymorphs of fergusonite.

F. *Samarskite*: (42) (*x*-ray data in Table 4)

All samarskite specimens examined were found to be metamict. Upon ignition the samples crystallized in a manner that depended largely on the method and temperature of ignition. Samarskite heated to ca 1000° C. in air gives consistently reproducible patterns for identification purposes.* When heated to approximately 1200° C. the specimens began to fuse, and the resultant powder patterns were poor and unreliable.

G. *Davidite*

The few samples studied were metamict. They crystallized readily when heated in air over a Meker burner or at higher controlled temperatures (to ca 1200° C.) to one structural type regardless of the temperature. The specimens examined give the same *x*-ray pattern as the Tete mineral described by Bannister and Horne (3).

H. *Allanite*: (*x*-ray data in Table 5)

Although allanite does occur fairly well crystallized, it very often is found in the metamict state. Some specimens may appear to be crystalline optically, having birefringence and giving an interference figure. However, this same material may give no diffraction pattern. Usually the specimens that do give a pattern have weak and diffuse peaks in the large 2θ region, indicating at least partial disorder.

When this mineral occurs in the metamict state it usually can be crystallized by heating to red heat in air or in an inert atmosphere. The resultant *x*-ray powder pattern is similar to that given by nonmetamict material. Although all nonmetamict allanites gave similar patterns, many showed significant differences in *d*-distances. Likewise, when crystalline allanite was ignited, the lattice parameters did not remain constant. Allanites fuse or decompose at relatively low temperatures so that, in order to get reliable *x*-ray patterns, the material must not be heated over ca 850° C.

* It is noted that the samarskite pattern in Table 4 does not correspond to that published by Murdock. However, because this pattern has been consistently reproduced for numerous samples of National Museum and other material labeled samarskite, it is believed this pattern is representative of that metamict mineral.

DISCUSSION

Data have been presented to aid in identifying some of the metamict minerals. This information is necessarily incomplete and may require subsequent revisions. It is believed, however, that enough new information has been obtained to serve as an aid to any future systematic work in this field.

In any study on these materials one must bear in mind that many of these metamict minerals have such complete internal disorder that they no longer fit the definition usually applied to the term "mineral," and that many have had certain constituents either added or subtracted since they were originally deposited. Hence, the term "recrystallization" is not strictly applicable in most instances, and "crystallization" would be more appropriate.

When metamict minerals are heated to comparatively low temperatures, structural changes take place at a slow rate. This sluggish crystallization becomes much more rapid with increase in temperature. Whereas crystallization of fergusonite may take place in less than 5 minutes at 800° C., the identical crystallization at 500° C. may take 24 hours. It has been observed that changes due to ignition are not confined to the metamict minerals. Data on the columbite-tantalite group (Table 6) and a barium-containing pyrochlore (Table 7) are included to show what happens to "nonmetamict" related minerals when subjected to elevated temperatures.

It is generally accepted (17, 23, 31 others) that the metamict condition is brought about by radiation from radioactive material included in the crystal, or from radioactive elements contained in the structure of the crystal. It is, however, a possibility that some of the massive varieties of metamict minerals may never have formed as crystalline material. Likewise, many of these minerals may have been stable only at the physical-chemical conditions at the time of formation; subsequent changes in these conditions, together with the included radioactivity, may have led to a rapid destruction of the metastable structures. This would explain why some of these minerals have never been found in a crystalline condition.

Data presented so far indicate that many metamict minerals may not recrystallize to the structure which they had when originally formed in nature. Hence, "reconstitution" data should be used with caution because such data in many instances may have no relation to the original structure of the mineral.

More work on the synthesis of those minerals that become metamict and artificial metamictization of these synthesized minerals would supply much valuable information on this problem.

ACKNOWLEDGMENTS

This work was begun while the author was with the U. S. Geological Survey, as part of the program being conducted on behalf of the U. S. Atomic Energy Commission. The methods and procedures were developed in the Survey's laboratories.

All of the final data presented in the tables were compiled from work conducted by the author as a member of the Bureau of Mines, U. S. Department of the Interior, at the Laboratories of the College Park, Md., station.

The author is especially indebted to George Switzer, U. S. National Museum, Washington, D. C., and to Clifford Frondel, Harvard University, for supplying many of the samples of metamict minerals.

TABLE 1. X-RAY DIFFRACTION DATA

Zircon* from Lincoln County, Calif. Not metamict		Cyrtolite(Hybla, Ont.) Not heated		Cyrtolite† (Hybla, Ont.) heated to ca 950° C. in air	
$d(\text{Å})^2$	$I.^1$	$d(\text{Å})$	$I.$	$d(\text{Å})$	$I.$
4.43	70	4.48VB	60	4.43	50
3.30	100	3.34VB	100	3.31	100
				2.97 ZrO ₂	40
2.65	10			2.65	5
2.52	60	2.54VB	30	2.52	60
		Traces ↓			
2.33	20			2.34B	10
2.22	15			2.22VB	5
2.06	40			2.07B	20
1.908	15			1.91B	15
				1.83B } ZrO ₂	10
				1.82B }	
1.751	20			1.754B	10
1.712	70			1.714	30
1.651	60			1.656	10
				1.555B ZrO ₂	10
1.547	5			1.548B	5

B = Broad peak, VB = Very broad.

* Tetragonal cell. $a_0 = 6.60 \pm .01^2$, $c_0 = 5.98 \pm .01$.

† Tetragonal cell. $a_0 = 6.62 \pm .03$, $c_0 = 5.98 \pm .03$.

¹ Intensities in all of the Tables are relative based on 100 for the strongest line.

² In this and all of the following tables lattice spacings and unit cell dimensions are in Ångstrom units. CuK α , wavelength of 1.5405 Å, is used to calculate the d values. The data have been obtained by a Norelco wide range goniometer using a silicon standard and having a 3° take-off. The goniometer was run at $\frac{1}{4}^\circ$ per minute.

TABLE 1—(continued)

Zircon* from Lincoln County, Calif. Not metamict		Cyrtolite (Hybla, Ont.) Not heated		Cyrtolite† (Hybla, Ont.) heated to ca 950° C. in air	
$d(\text{Å})^2$	<i>I</i> ¹	$d(\text{Å})$	<i>I</i> .	$d(\text{Å})$	<i>I</i> .
1.494	1			1.50B	5
1.476	25			1.483B	10
1.381	20			1.385B	5
1.362	7			1.365B	5
1.290	7			1.289VB	1
1.259	5			1.259VB	3
1.249	5				
1.246	3				
				1.191B	5
1.188	20			1.186B	1
1.167	5				
1.109	5			1.109VB	1
1.101	23				
1.068	3				
1.059	15			1.054VB	5
1.051	5				
1.048	5				
1.045	5				
1.042	3				
1.002	2				
0.9751	2			0.976VVB	1
.9719	3			Broad	
.9538	3			Traces	
.9321	5				
.9200	2				
.9160	3				
.8999	3				
.8975	3			.897VVB	1
.8919	10				
.8867	5				
.8829	2				
.8562	5			.857VVB	3
.8530	2				
.8398	2				
.8331	10				
.8256	5				
.8118	5				
.8011	7				
.7901B	7				
.7811	10				

TABLE 2

Thorite* partly metamict from Nigeria		Thorite† heated to ca 1050° C. in air Same material from Nigeria		Uranothorite‡ (Mostly metamict) heated to ca 1300° C. in air. Locality unknown	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
4.68	100	4.69	80	5.25	20
				4.71	10
				4.65	50
				4.18	90
				4.03	40
3.55	100	3.53	100	3.56	10
				3.49	40
		3.34(SiO ₂)	5	3.27	60
		3.18(ThO ₂)	20	3.13B(U, ThO ₂)	2
				3.07	100
				2.96	30
				2.88	10
2.84	20	2.83	20	2.86	70
2.82		2.82			
2.65B	60	2.66	60	2.70B(U, ThO ₂)	2
				2.67	5
				2.63	20
		2.51	20	2.51	2
2.50B	15			2.46	20
				2.42	15
				2.38	5
				2.34	5
2.22B	20	2.21	20	2.17	20
				2.16	20
				2.14	25
				2.10	20
2.00B	20	2.005	10	1.94B(U, ThO ₂ ?)	25
		1.94B(U, ThO ₂ ?)	1	1.914	10
				1.891	15
1.88B	20	1.877B	10	1.870	20
		1.865B			
		1.823	30	1.847	15
1.825B	30				

B=Broad peak.

* Tetragonal cell. $a_0=7.10 \pm .02$, $c_0=6.32 \pm .02$.† Tetragonal cell. $a_0=7.05 \pm .02$, $c_0=6.32 \pm .02$.‡ Monoclinic cell. Using Pabst's value for $\beta=104^\circ 55'(45)$ this pattern gives: $a_0=6.74 \pm .02$, $b_0=6.95 \pm .02$, $c_0=6.60 \pm .02$.

TABLE 2—(continued)

Thorite*-partly metamict from Nigeria		Thorite† heated to ca 1050° C. in air. Same material from Nigeria		Uranothorite‡ (Mostly metamict) heated to ca 1300° C. in air. Locality unknown	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
				1.793	10
				1.770	15
		1.760B			
				1.754B	2
				1.740	15
				1.720	20
				1.679	5
		1.660B	10		
				1.633B	10
				1.589B	10
		1.580B	10		
				1.568B	2
				1.540B	10
		1.480B	10	Traces	
		1.437}	10		
		1.433}	10		
		1.330B	10		
				1.369B	10
				1.330B	10
				1.323B	10
				1.292}	10
				1.289}	10
				1.283B	2
		1.270B	2	1.273B	5
		1.170B	2	Traces	
		1.120B	2		
				1.078B	5
		1.060B	2	1.060B	10
		1.02B	2	1.030B	5
				.9678B	5
				.9563B	10
				.8629B	5
				.8166B	5
				.8075B	5
				.8070B	5
				.7894B	5

TABLE 3

Euxenite (No. BM 26)* heated to ca 1200° C. in air		Euxenite (No. BM 26) heated to ca 430° C. in air		Euxenite (No. BM 26) heated to 1250° C. in helium	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
7.28	15	5.47	20		
Weak		4.62	7		
broad peaks		4.46	7		
3.66	40	4.41	20	3.67	1
3.37	20	4.25	10		
3.28	10	3.96	10	3.30	40
				3.13	60
2.99	100	3.76	5	2.99B	1
2.95	40	3.70	7	2.96	100
2.78	25	3.34B	10	2.75	30
2.63	10	3.07	30	2.65	20
2.60	30	2.98	70		
2.55	20	2.91	100		
2.52	5	2.77	20	2.53	40
2.45+}	15	2.59B	20		
2.45-}		2.43	5		
2.43	20	2.23	20		
2.31	17	2.21	10	2.34	10
2.21	13	2.11B	5	2.23	10
2.19	13	2.03B	7	2.17	10
2.12	20	1.980	20		
2.08	3	1.950	10	Faint	
2.04	5	Weak		Traces	
1.979	10	Peaks		1.98	1
1.941}	13	1.855	20		
1.939}		1.820B	10		
1.897	25	1.732B	5	1.91	50
1.880B	3	1.681	15	1.87	30
1.830	30	1.676	15	1.84B	1
1.806	20	1.638B	5	1.81	60
1.774	25	1.576	15	1.77B	2
1.731	25	1.563B	5		
1.727	27	1.505VB	10		
1.704	3	Weak		1.71	40
1.683	3	Peaks			
				1.655	30
				1.651	20
1.643	25	1.195B	10		
1.625	5	1.185B	3		
1.614B	10	1.166B	5		
1.590	11	1.150B	3		
1.565	15	1.148B	3	1.572B	20
1.540	15	1.141B	2	1.544	60
1.505	10	1.128B	3	1.511B	10

B = Broad peak, VB = Very broad.

* Sample from Risö, Norway; very similar patterns are given by euxenites from Kragerö, Norway; Lyndoch, Ontario; Perth, Ontario; Swaziland, South Africa; Spruce Pine, N. C.; Iveland, Norway; Hiterö, Norway; and other unknown localities.

TABLE 3—(continued)

Euxenite (No. BM 26)* heated to ca 1200° C. in air		Euxenite (No. BM 26) heated to ca 430° C. in air		Euxenite (No. BM 26) heated to 1250° C. in helium	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
1.496	15	1.095B	3		
1.492	25	1.069B	3		
1.488		1.061B	3		
1.476		1.054	3	1.478	20
1.474	5	Weak peaks			
1.462	7	.943B	3		
1.441	10	.886B	3	1.385	20
1.437	10			1.369	10
Weak peaks					
1.344	5				
1.336	5				
				1.326B	3
1.296	10				
1.278	3			1.280B	3
1.277					
1.246	3			1.262B	3
1.224B	3			1.243B	1
1.209B	3			1.224B	10
1.187B	5			1.195B	10
1.173B	5				
1.160	7			1.174B	5
1.157					
1.143B	3			1.158B	3
1.119B	3			1.142B	3
1.102B	3			1.114B	10
1.082B	3				
1.058B	3			1.086B	2
1.056B	3			1.060B	3
1.043B	3			1.053B	3
1.039B	2			1.046B	20
1.034B	3				
1.0235B	3				
1.0205B	3				
1.0170B	3				
.9991B	2				
.9966B	2				
.9892B	2				
.9832B	3			.9842B	10
.9630B	3				
.9430B	2				
.9155B	5				
.9104B	2				
				.9058B	10
.8626B	2			.8958B	2
.8592B	2			.8652B	10

TABLE 4

Fergusonite* (No. LM-1) ² heated to ca 1200° C. in air		Fergusonite (No. LM-1) ¹ heated to ca 700° C. in air		Samarskite† heated to ca 1050° C. in air ³	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
5.47	3	3.04	100		
4.58	3	2.74	30		
4.02	3	2.59VB	30	4.03	20
		1.88B	40	3.59	20
3.29	3	1.63	10	3.23B	30
3.12	100	1.57}B	5	3.13	40
3.01	20	1.56}	2		
2.96	90	1.22B	2	2.98	100
				2.92	90
				2.81B	5
2.74	40			2.75	10
2.64	20			2.64B	5
2.59B	2			2.59	5
2.53B	10			2.52B	20
2.52	10				
				2.45	10
2.22	3			2.33	5
2.16	5			2.17B	5
				2.07B	2
2.01	5			2.05B	3
1.901	50			1.909	20
				1.890B	10
				1.863	20
1.855	30				
				1.830	20
1.754B	10				
				1.739	10
				1.709	20
				1.689	20
				1.654	
1.646	10			1.650	5
1.643B	10				

B=Broad peak, VB=Very broad.

* Sample supplied by Louis Moyd from Madawaska, Ontario. Nearly identical patterns are given by fergusonite from: Ahikambara, Madagascar; Llano Co., Texas; Amherst Co., Va. (2 localities); Arendal, Norway; Hiterö, Norway; Mitchell Co., N. C.; and other unknown localities.

† Locality unknown. Nearly identical patterns are given by samarskite from: Wheatland, Wyo.; Glastonbury, Conn.; Molinda, Ga.; Mitchell Co., N. C. and other localities.

¹ Tetragonal cell. $a_0=5.18 \pm .02$, $c_0=5.48 \pm .02$.

² Tetragonal cell. Unit cell as indexed by Barth. $a_0=7.74$, $c_0=11.34$.

³ Could not be indexed.

TABLE 4—(continued)

Fergusonite* (No. LM-1) ² heated to ca 1200° C. in air		Fergusonite (No. LM-1) ¹ heated to ca 700° C. in air		Samarskite† heated to ca 1050° C. in air ³	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
1.627B	15			1.630VB	5
1.620	15				
1.569	10			1.573B	10
1.563B	15			1.561	30
1.559	10				
1.508B	5			1.515B	10
1.504	5				
1.500B	5			1.500B	5
1.478B	3			1.478B	3
				1.462B	5
				1.437B	10
				1.425B	5
1.369	3				
1.321B	2				
1.288B	1				
1.263B	1				
1.245B	1				
1.236B	2				
				1.221B	1
1.213VB	3				
1.189	3			1.188B	1
1.146VB	3				
1.107B	3				
1.088B	2				
1.082B	3				
1.079B	3				
1.053B	1			1.056VB	1
1.048B	2				
1.046B	3				
1.041B	2				
1.036B	2				
1.025B	1				
1.001B	3				
0.9856B	1				
0.9168B	2				
0.9112B	2				
Traces					

TABLE 5

Allanite-mostly metamict (No. H. M. 86077)		Allanite-not metamict (supplied by Brian Mason)		Metamict allanite* (No. H. M. 86077) heated to ca 800° C. in air ½ hr.		Metamict allanite (No. H. M. 86077) heated to ca 850° C. for 14 hr.	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
		9.20	20			9.30	30
		8.05	20				
		7.96	20				
		5.09	10	5.09	10	5.07	30
		5.01	10				
		4.87	5			4.84	10
		4.69	20	4.65	30	4.67	10
						4.62	50
						4.43B	2
						4.10B	2
						4.00	20
		3.78	10	3.79B	2	3.81B	10
		3.59	10	3.59B	1	3.60B	1
3.52VB	50	3.52	40	3.51	80	3.50	80
						3.34	30
		3.30	10	3.28B	2	3.27	20
		3.23	20	3.22B	2	3.20	10
				2.95B	100	2.96	100
2.93VB	100	2.92	90				
		2.91	100				
		2.86	50	2.84B	40		
		2.82	10			2.83	30
						2.79B	2
						2.74	30
2.69VB	30	2.68	40	2.68B	60	2.67	80
2.62VB	10	2.62	40	2.61B	50	2.60	40
		2.51	20			2.54	30
		2.50B	10			2.48B	2
						2.43	30
						2.40B	2
		2.33B	10			2.33B	10
						2.24B	10
		2.20	10			2.20B	10
		2.18B	20	2.16B	2	2.16B	30
		2.15	10			2.13B	30
		2.14	10				
		2.10B	2				
		2.05B	2			2.06B	10

B=Broad peak, VB=Very broad.

* Similar patterns given by samples from Arendal, Norway (orthite); Johannesburg, So. Africa, and numerous other unlisted localities.

TABLE 5—(continued)

Allanite-mostly metamict (No. H. M. 86077)		Allanite-not metamict (supplied by Brian Mason)		Metamict allanite* (No. H. M. 86077) heated to ca 800° C. in air ½ hr.		Metamict allanite (No. H. M. 86077) heated to ca 850° C. for 14 hr.	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
		1.91B	2			1.90B	20
		1.89B	20				
		1.78B	2				
		1.67B	10				
		1.65B	10			1.65B	30
		1.64B	20			1.63B	60
		1.56B	20				
						1.47VB	10
						1.42B	20
		1.12B	2				

TABLE 6

Columbite #1*		Columbite #1† heated to ca 1250° C. in air		Columbite-Tantalite‡ (No. Ta 14)-high tantalum	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
7.15	1	7.15	20	7.15	5
		5.33	10	5.33	3
		5.15	10		
		4.78	10		
		3.74*	20		
3.65	40	3.67	50	3.67	60
		3.60*	30		
		3.57	25	3.58	5
		3.03	10		
2.97	100	2.97	100	2.98	100
		2.93*	50		
2.87	20	2.87	10	2.87	10
		2.86	10		
		2.83	5		
		2.82*	5		
		2.78	10		
		2.77	5		
2.55	10	2.54	10	2.55	7
		2.53	5		

* = FeNb₂O₆.

B = Broad Peak.

* Orthorhombic cell. $a_0 = 5.10 \pm .01$, $b_0 = 14.24 \pm .01$, $c_0 = 5.73 \pm .01$.

† Mitchell Co., N. C.

‡ Black Hills, South Dakota.

† More than one compound.

‡ Orthorhombic cell. $a_0 = 5.10 \pm .01$, $b_0 = 14.24 \pm .01$, $c_0 = 5.73 \pm .01$.

TABLE 6—(continued)

Columbite #1*		Columbite #1† heated to ca 1250° C. in air		Columbite-Tantalite ^{2†} (No. Ta 14)-high tantalum	
<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>	<i>d</i> (Å)	<i>I.</i>
2.50	20	2.50	25	2.50	20
		2.47	10		
		2.43	5		
2.38B	2	2.39	10	2.38B	7
		2.34	5		
		2.24	3	2.25	3
2.21	15	2.21	10	2.22	7
		2.18	10		
2.09	10	2.09	10	2.09	7
		2.06	10		
		2.05	10		
		2.00	10		
1.91	20	1.907	10	1.91	7
1.83B	10	1.834	15	1.836	7
		1.805	5		
1.77B	10	1.780	20	1.777	15
		1.773	30		
1.74B	20	1.740B	20	1.745	20
		1.737	40		
1.72B	30	1.726	40	1.727	30
		1.698	20		
		1.580	10		
		1.573	10		
		1.562	10		
1.542B	20	1.532	20	1.542	10
		1.487	10	1.490	7
1.464B	30	1.467	20	1.466	15
		1.456	20	1.459	20
		1.440	20		
1.380B	10	Traces		1.384	3
Traces ↓		1.244	10	1.248	2
		1.224	10	1.225}	3
				1.223}	
		1.198	20	1.196	7
		1.179	10		
				1.140	3
		1.136	5		
		1.121	5		
		1.103	10	1.103	5
				1.092B	2
		1.080B	5	1.079B	2
				1.067B	1
				1.048B	2
		1.032B	5	1.036B	2
		1.020B	5	1.024B	3
		Traces		.997B	3
				.925B	2
				.883B	2
				.840B	2

TABLE 7

Barium-bearing pyrochlore*		Sample heated to ca 900° C. in air	
$d(\text{Å})$	$I.$	$d(\text{Å})$	$I.$
6.08	80	7.14	5
		5.97	20
		5.34B	2
		3.87	80
		3.59B	1
3.18	40	3.19	10
		3.13	100
3.04	100	3.01	60
		2.77	20
2.64	30		
		2.59	10
2.42	10	2.44	50
		2.39	10
		Traces	
2.03	10	2.00B	2
		1.940B	10
1.86	30	1.860B	1
		1.838B	10
		1.808B	10
1.78	10		
		1.763B	5
		1.642B	20
1.61B	2		
		1.609}	2
		1.602}	
1.590	30		
		1.568B	10
1.541B	1		
1.521B	10		
		1.499B	1
1.478B	5		
		1.456B	5
		Traces	
1.374B	1		
		1.326B	
1.319B	1	1.320B	2
1.210B	10		
1.180B	5		
1.158B	2	1.182B	5

B=Broad peak, VB=Very broad.

* Sample from Alaska; exact locality unknown.

$a_0 = 11.53 \pm .02$.

TABLE 7—(continued)

Barium-bearing pyrochlore*		Sample heated to ca 900° C. in air	
<i>d</i> (Å)	<i>I</i> .	<i>d</i> (Å)	<i>I</i> .
1.076B	5		
1.016	10		
Traces			
.893VB	10		
.881VB	5		
.836VB	2		

REFERENCES

- *1. ALLEN, A. O., AND GHORMLEY, J. A. (1947), Decomposition of solid barium nitrate by fast electrons: *Jour. Chem. Phys.*, **15**, 208–209.
2. ARNOTT, RONALD J. (1950), X-ray diffraction data on some radioactive oxide minerals: *Am. Mineral.*, **35**, 386–400.
3. BANNISTER, F. A., AND HORNE, J. E. T. (1950), A radioactive mineral from Mozambique related to davidite: *Mineral. Mag.*, **29**, 101–112.
- *4. BARTH, TOM F. W. (1926), The structure of synthetic, metamict, and recrystallized fergusonite—The structure of Risörite: *Norsk. geol. Tidsskr.*, **9**, 23–39.
5. BERMAN, JOSEPH (1951 Abstr.), Studies of metamict minerals. (I): Methods and procedures: *Geol. Soc. Am., Bull.* **62**, 1422–1423.
6. ——— (1952 Abstr.), Studies of metamict minerals (II): Re-examination of fergusonite: *Geol. Soc. Am., Bull.* **63**, 1235.
7. BOROVSKII, I. B., AND BLOKHIN, M. A. (1931), Roentgen study of metamictic state of zircon: *Contr. of Lomonosov Inst., Ac. of Sci.*, **7**, 197–207.
8. BONATTI, S., AND GALLITELLI, P. (issued 1951; dated 1950), Sulla Torite di Nettuno (Roma): *Att. soc. tosc., Mem.*, **57**, (A) 3–4.
9. BRANDT, KARIN (1943), X-ray studies on ABO_4 compounds of rutile type and AB_2O_6 compounds of columbite type: *Arkiv. För Kemi, Mineralogi Och Geologi, Band 17 A*, No. 15, 1–8.
10. BROOKER, E. J., AND NUFFIELD, E. W. (1951 Abstr.), Studies of radioactive compounds: IV—Pitchblende from Lake Athabasca: *Geol. Soc. Am., Bull.* **62**, 1425–1426.
- *11. ——— (1952), Studies of radioactive compounds: IV—Pitchblende from Lake Athabaska, Canada; *Am. Mineral.*, **37**, 363–385.
- *12. BUERGER, M. J. (1948), The role of temperature in mineralogy: *Am. Mineral.*, **33**, 101–121.
- *13. ——— (1951), Crystallographic Aspects of Phase Transformations: Common Solids, Div. of Phy. Sci. Nat. Research Council, Symposium at Cornell U. (Aug. 1948) on Phase transformations in solids—John Wiley and Sons, Inc., 183–211.
14. CHUDOBA, K. F., AND LANGE, H. (1949), Rekrystallisationsversuche an autoisotropisiertem Gadolinit, Samarskit und Euxenite: *Neues Jb. Min., Monatshefte, A*, 30–45.
15. CHUDOBA, K., AND V. STACKELBERG, M. (1936), Dichte und Struktur des Zirkons: *Zeit. Krist.*, **95**, 230–246.
16. DAMOUR, M. A. (1864), Note sur la densité des zircons: *Compt. Rend.*, **58**, 154–159.

- *17. DANA, J. D., AND E. S. (1944), The System of Mineralogy, ed. 7, vol. 1, by C. Palache, H. Berman, and C. Frondel, John Wiley and Sons, Inc., N. Y.
18. DES CLOIZEAUX, A., AND DAMOUR, A. (1806), Examen des proprietes optiques et pyrogenetiques des mineraux connus sous les noms de gadolinites, allanites, orthites, euxenite, tyrite, yttrorantalite et fergusonite: *Ann. chim. phys.* (3), **59**, 357-379.
19. DIHLSTRÖM, KLAS (1938), Über den Bau des wahren Antimontetroxyds und des damit isomorphen Stibiotantalits, $SbTaO_4$: *Zeit. anorganische und allgemeine Chemie*, Band **239**, 57-64.
20. ELLSWORTH, H. V. (1925), Radioactive minerals as geological age indicators: *Am. J. Sc.*, **209**, 127-144.
21. ——— (1932), Rare element minerals of Canada: *Geol. Surv. Canada, Econ. Geol. Series II*.
22. EVANS, R. D., AND GOODMAN, C. (1941), Radioactivity of rocks: *Geol. Soc. Am., Bull.* **52**, 459-490.
- *23. FAESSLER, A. (1942), Untersuchungen zum Problem des metamikten Zustandes: *Zeit. Krist.*, **104**, 81-113.
24. GEORGE, D. R. (1951), Thorite from California: *Am. Mineral.*, **36**, 129-132.
- *25. GOLDSCHMIDT, V. M. (1924), Über die Umwandlung krystallisierter Mineralien in den metamikten Zustand (isotropisierung): *Norsk. Ak., Skr.*, L. 1. No. 5, Geochemische Verteilungsgesetze der Elemente. III. (Anhang), 51-58.
26. HAMBERG, AXEL (1914), Die radiaktiven Substanzen und die geologische Forschung: *Geol. För. Förh.*, **36**, 31-96.
27. HESS, F. L., AND WELLS, R. C. (1930), Samarskite from Petaca, New Mexico: *Am. J. Sc.*, **19**, 18-26.
- *28. HOLLAND, HEINRICH D., AND KULP, LAURENCE J. (1950), Geologic age from metamict minerals: *Science*, **3**, 312.
29. HURLEY, PATRICK M. (1951), Alpha ionization damage as a cause of low helium ratios: *Technical Report, Office of Naval Research, Contract No. N5ori-07829*.
30. ———, AND FAIRBAIRN, H. W. (1952), Alpha-Radiation damage in zircon: *Jour. Appl. Phys.*, **23**, No. 12, 1408.
- *31. ——— (1953), Radiation damage in zircon: A possible age method, *Geol. Soc. Am., Bull.* **64**, 659-673.
- *32. HUTTON, C. OSBORNE (1950), Studies of heavy detrital minerals: *Geol. Soc. Am., Bull.* **61**, 635-716.
- *33. KOSTYLEVA, E. E. (1951), On metamictic disintegration of the zircon group of minerals—Problems of mineralogy, geochemistry and petrography, *Ac. Sc. U.S.S.R.*, 27-35 (in Russian).
- *34. KULP, J. L., VOLCHOK, H. L., AND HOLLAND, H. D. (1952), Age from metamict minerals, *Am. Mineral.*, **37**, 709-718.
35. LARSEN, E. S., JR., WARING, C. L., AND BERMAN, J. (1953), Zoned zircon from Oklahoma: *Am. Mineral.*, **38**, 1118-1125.
36. LIEBISCH, TH. (1910), Über die Rückbildung des kristallisierten Zustandes aus dem amorphen Zustande beim Erhitzen pyrognomischer Mineralien: *Ak. Berlin Ber.*, **I**, 350-364.
37. MACHATSCHKI, F. (1929), Über die Formel des Risörites und Fergusonites: *Zeit. Krist.* **72**, 291-300.
- *38. ——— (1941), Zur Frage der Stabilität des Zirkongitters: *Zbl. Min.*, **A**, 38-40.
- *39. MOREHEAD, F. F., AND DANIELS, F. (1952), Storage of radiation energy in crystalline lithium fluoride and metamict minerals, *Jour. Phys. Chem.*, **46**, 546-548.

40. MORGAN, J. H., AND AUER, MARIANNA L. (1941), Optical, spectroscopic and radio-activity studies of zircon: *Am. J. Sc.*, **239**, 305-311.
41. MÜGGE, O. (1922), Über isotrop gewordene Kristalle: *Cbl. Min.*, 721-739 and 753-765.
42. MURDOCH, JOSEPH (1951), Notes on some California minerals: *Am. Mineral.*, **36**, 358-359.
43. ORCEL, JEAN (1953), Analyse thermique differentielle de quelques minéraux métamictes: *Compt. Rend.*, **236**, 1052-1054.
44. ORCEL, JEAN ET LÉVY, CLAUDE (1953), Analyse thermique de la betafite, mineral métamictic: *Compt. Rend.*, **236**, 1, 1177-1179.
45. PABST, A., AND HUTTON, C. OSBORNE (1951), Huttonite, a new monoclinic thorium silicate; with an account of its occurrence, analysis and properties: *Am. Mineral.*, **36**, 60-69.
46. PABST, A. (1951), X-ray examination of uranorthorite: *Am. Mineral.*, **36**, 557-562; abstract in *Geol. Soc. Am., Bull.* **61**, 1492 (1950)
- *47. ——— (1952), The metamict state: *Am. Mineral.*, **37**, 137-157.
48. ——— (1954), Brannerite from California: *Am. Mineral.*, **39**, 109-117.
- *49. PELLAS, P. (1951), Mineralogie—sur la destruction spontanée des réseaux cristallins de minéraux radioactifs: *Compt. Rend.*, **232**, 1369-1371.
- *50. ——— (1953), Sur l'établissement de l'état métamictic dans gadolinite. Bilan énergétique de la recristallization: *Compt. Rend.*, **236**, 619-621.
51. PETERSON, WALFR. (1890), Studier öfver gadolinit: *Geol. För. Förh.*, **12**, 275-347.
52. PRIOR, G. T. (1894), On fergusonite from Ceylon: *Mineral. Mag.*, **10**, 234-237.
- *53. SEITZ, F. (1949), On the disordering of solids by action of fast massive particles: *Proc. Faraday Soc.*, 271-282.
- *54. SLATER, J. C. (1951), The effects of radiation on materials: *Jour. Appl. Phys.*, **22**, 237-256.
55. V. STACKELBERG, M., AND CHUDOBA, K. (1937), Dichte und Struktur des Zirkons, II: *Zeit. Krist.*, **97**, 252-262.
56. V. STACKELBERG, M., AND ROTTENBACH, E. (1940), Dichte und Struktur des Zirkons, III and IV: *Zeit. Krist.*, **97**, 173-182 and 207-208.
- *57. STECH, B. (1952), Structural change produced in crystals by alpha particle bombardment: *Zeit. Naturforsch.* **7a**, Feb. 175-185.
58. VEGARD, L. (1916), Results of crystal analysis: *Phil. Mag.* (6), **32**, 65-96.
59. ——— (1927), Results of crystal analysis: *Phil. Mag.* (7), **4**, 11.
60. VOGT, TH. (1911), Vorläufige Mitteilung über Yttrfluorite, eine . . . : (On non-metamictic fergusonite): *Centralblatt für Min., Geologie und Paläontologie*, **12**, 373.
61. WYCKOFF, RALPH W. G., AND HENDRICKS, STERLING B. (1927), Die Kristallstruktur von Zirkon und die Kriterien für spezielle Lagen in tetragonalen Raumgruppen: *Zeit. Krist.*, **66**, 73-102.
62. ZHIROV, K. K. (1952), Conversion of zircon into the metamict state: *Doklady Akad. Nauk., U.S.S.R.*, **85**, 889-891 (in Russian).

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