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URANIUM AND THORIUM IN THE ACCESSORY ALLANITE OF IGNEOUS ROCKS*

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By William Lee Smith, Mona L. Franck,
and Alexander M. Sherwood

ABSTRACT

Accessory allanite was separated from phanerocrystalline igneous rocks and its optical properties and radioactive components were compared. The indices of refraction of these allanite samples are higher than those from the pegmatites that are usually described in geologic literature. The birefringence was found to range from 0.015 to 0.021, the α index of refraction from 1.690 to 1.775. The allanite content ranged from 0.005 to 0.25 percent by weight in the rocks studied. The mineral is confined largely to the more siliceous phanerites. The uranium content is highest in the allanite from granites, regardless of area, ranging from 0.004 to 0.066 percent, whereas the thorium content is high or low regionally, ranging from 0.35 to 2.33 percent. Allanite was found to be otherwise of exceptionally uniform composition.

INTRODUCTION

Allanite, $(RE,^{*} Ca)_2(Fe,Al)_3Si_3O_{12}(OH)$, may occur as an accessory mineral in siliceous and intermediate igneous rocks, in limestone contact skarns, pegmatites, crystalline metamorphic rocks, and as a component of magmatic iron ores. The mineral is monoclinic and varies in color from light brown to black. Its hardness ranges from 5 to 6 depend-

*Rare earths

ing upon its degree of alteration, and similarly its specific gravity varies from 3.4 to 4.2. Allanite is a member of the epidote group with rare earths substituting for calcium. Allanite is often found with epidote; some of it is intergrown with epidote (Hobbs, 1889). The metamictization of allanite produces an amorphous alteration product, and some allanite from pegmatite is completely isotropic. The alteration is inferred to be the result of the destruction of the crystalline structure of allanite by the radioactive decay of its uranium and thorium.

MINERALOGY

Separation methods

In the granitic rocks that were studied, allanite was found to average 0.1 mm in diameter. The rocks were pulverized on a rolls-type crusher which freed the allanite cleanly from its hosts. The allanite was generally found to concentrate in the 100- to 200-mesh size fraction. Allanite concentrates with the other accessory minerals in the sink of a methylene iodide separation (sp. gr. 3.33). It is easily separable from the other heavy minerals on a Frantz Isodynamic separator, allanite becoming magnetic between 0.4 and 0.6 amp at cross and longitudinal settings of 10°. When epidote occurs with allanite, the separation becomes more difficult. Methylene iodide saturated with iodoform (sp. gr. 3.45) was found to float epidote and sink the allanite in the concentrates studied. Thus a minimum of hand-picking was required to obtain clean separates of the minerals.

Table 1.--Indices of refraction of allanite from igneous rocks.

Host rock	α	β	γ	Birefringence
Southern California batholith				
Coarse phase Rubidoux granite, Riverside	1.775	1.789	1.791	0.016
Fine phase Rubidoux granite, Riverside	1.735	1.750	1.752	0.017
Woodson granodiorite, Descanso	1.745	1.760	1.763	0.018
Woodson granodiorite, Temecula	1.735	1.750	1.753	0.018
Woodson granodiorite, Rainbow	1.740	1.755	1.759	0.019
Woodson granodiorite, Elsinore	1.740	1.755	1.760	0.020
Stonewall granodiorite, Cuyamaca	1.705	1.717	1.720	0.015
Mount Hole granodiorite, Mount Hole	1.695	1.710	1.714	0.019
Aguanga tonalite, Aguanga	1.743	1.760	1.763	0.020
Sierra Nevada batholith				
Basin Mountain quartz monzonite	1.750	1.766	1.770	0.020
Idaho batholith				
Porphyritic granodiorite, Cascade	1.740	1.751	1.755	0.015
Granodiorite, Stanley	1.761	1.776	1.780	0.019
Granodiorite, Atlanta	1.752	1.768	1.771	0.019
White Mountains, New Hampshire				
Fresh Conway granite, Conway	1.721	1.738	1.742	0.021
Weathered Conway granite, Conway	1.720	1.737	1.740	0.020
Albany quartz syenite, Passaconaway	1.690	1.700	1.706	0.016

CHEMISTRY

Chemical analysis

Chemical analysis (table 2) of allanite from different areas and of different optical properties were made by Glen Edgington of the Geological Survey.

Table 2.--Chemical analyses, in percent, of allanite from Cascade, Idaho; Conway, New Hampshire; and Riverside, California.1/

Constituent	Cascade granodiorite	Conway granite	Rubidoux granite (fine), Calif.
SiO_2	30.35	26.05	29.75
Al_2O_3	7.56	7.54	8.42
Fe_2O_3	18.14	17.01	20.68
CaO	12.90	10.45	8.95
MgO	1.43	0.81	0.91
MnO	0.38	0.58	0.31
H_2O (total)	2.00	5.60	2.40
Ce_2O_3	11.06	12.45	11.38
RE_2O_3 (other)	15.69	18.69	15.81
(Total RE incl. ThO_2)	(26.75)	(31.14)	(27.19)
Total	99.51	99.18	98.61
Determined on separate samples by Sherwood 2/			
ThO_2	1.2	0.92	0.76
U	0.0036	0.0540	0.0400

1/ Analyses made by classical methods outlined in Hillebrand and Lundell (1929).

2/ Uranium determined fluorimetrically. Thoria determined colorimetrically by thoron method.

Spectroscopy

Semiquantitative spectrographic analyses of nine allanites from igneous rocks are compared in table 3. The minerals were separated from rocks of the White Mountains, N. H., batholith, the Southern California batholith, the Sierra Nevada batholith, and the Idaho batholith. The rocks include fresh and weathered alkaline biotite granites, a hypersthene-bearing leucogranite, four granodiorites, a quartz monzonite, and a porphyritic granodiorite. Despite variations in the optical properties, color, abundance, and origin of these allanite specimens, they are remarkably alike in composition.

Aside from the radioactive components, which are discussed separately, most of the variations in composition are within the limits of precision of the method. One immediately obvious fact is that each allanite contains the same 37 elements, with the exception of the absence of Tm from the Basin Mountain mineral and the absence of Lu from the Cascade granodiorite mineral. The variations, which seem to be regional, are the higher content of Nb, Be, and Sn in the New Hampshire minerals, and the higher content of the elements Mn, Ti, Ni, and Cu in the Sierra Nevada specimen.

Among the rare earths, in all samples the order of abundance is Ce,(La,Nd,Pr), (Sm,Gd), variable traces of Lu, Ho, and Eu. Tm is the least abundant. Of the cerium earths the order is Ce,(La,Nd,Pr), Sm, and Eu. Of the yttrium earths the order is (Gd,Dy), (Er,Yb,Lu), (Ho,Tm.)

This distribution of rare earths shows the most pronounced cerium assemblage (Rankama and Sahama, 1950) of any of the rare-earth minerals. Goldschmidt and Thomassen (1924) describe six assemblages of rare earths occurring in minerals including an allanite-type assemblage containing the

Table 3.--Semiquantitative spectrographic analyses of allanite. Analyst: Mona L. Franck

Rock	Fresh Conway granite	Weathered Conway granite	Rubidoux granite, fine phase	Woodson grano-diorite	Woodson grano-diorite	Stonewall grano-diorite	Quartz monzonite	Porphyritic granodiorite	Granodiorite
Location	Conway, N. H.	Conway, N. H.	Riverside, Calif.	Temecula, Calif.	Descanso, Calif.	Cuyamaca, Calif.	Basin Mt., Calif.	Cascade, Idaho	Atlanta, Idaho
Percent									
>10.	Si Fe	Si Fe	Si Fe	Si Fe	Si Fe	Si Fe	Si Fe	Si Fe	Si Fe
5-10.	Ce Ca	Ce	Ce Ca Al	Ce	Ce Al Ca	Ce Al Ca	Ce Al	Ce Al Ca	Ce Al Ca
1- 5.	Al La Nd Pr	Al Ca La Nd Th Pr	La Nd Pr	Al Ca La Nd Pr	Nd La Pr	Nd La Pr	Ca La Pr Nd	La Nd Th Pr	Nd La Pr Th
0.5- 1.	Th Y Mg	Y Mg	Th Y Mg	Mg Y Th	Y Mg Th	Th Y Mg	Mg Ti Mn	Mg	Y Mg
0.1- 0.5	Mn Ti Sm Zr Gd	Zr Ti Sm Mn Gd	Zr Ti Mn Sm Gd	Ti Mn Sm	Ti Zr Mn Sm Gd	Ti Mn Sm Gd	Th Y	Ti Mn Y Sm	Mn Gd Ti Sm
0.05- 0.1	Dy Sr Er Yb	Dy Er Yb Sr	Dy Er Yb	Gd Dy Er	Dy Er B Yb	Er Zr Dy Sr	Sm Zr Yb Dy Gd	Sr Zr Gd	Dy Sr Er
0.01- 0.05	Ba Nb Pb V Sc Lu Sn Ho	Nb Ba V Pb Sc Sn Lu Ho	Sr V Pb Sc Ba Ho Lu	Zr Sr V Yb Ba Sc Pb	Sr Ba Pb V Sc Ho Lu	V Yb Sc Ba B Ho Lu Pb	Er Ba Sr Pb Sc V Cu Ni Nb	Dy Er V Sc Ba Pb Yb	Yb Zr V Sc Lu Pb Ho
0.005- 0.01	B Eu Ga Ni	B Eu Ni Ga	B Eu Gd Ni	Ho B Eu Ga Lu Ni	Eu Ni Ga Cu	Eu Ni Ga Cu	Eu B Ga Lu Ho	Eu B Ga Ni	Nb Ba Eu B Ga Ni
0.001- 0.005	Cr Be Cu Co Tm	Cr Cu Be Co Tm	Cr Nb Cu Co Sn Tm	Cr Nb Tm Co Cu Sn	Cr Nb Co Sn Tm	Cr Nb Tm Co Sn	Cr Co Sn	Ho Cr Cu Co Sn Nb Tm	Sn Cr Cu Co Tm
0.0005- 0.001			Be		Be				Be
0.0001- 0.0005				Be		Be	Be	Be	

series La-Nd, minor amounts of Sm-Gd, and traces of other yttrium earths.

The sensitivity of the spectrographic method used for Na is 0.01 percent, for K is 0.3 percent, and for Li is 0.04 percent. The most sensitive lines were not used for these analyses because their wavelengths are in the visible region of the spectra and the standard method covers from 2350 A to 4750 A.

Radioactive components

Table 4 compares the Th and U contents, calculated eU, Th/U, and optical properties of ten of the allanite samples. This radioactivity, called calculated total eU in table 4, is calculated (for beta counting), on the assumption of the secular equilibrium of both U and Th, by taking the sum of the percent U and one-quarter of the percent Th. The minerals contain no detectable potassium.

In the periodic system uranium and thorium belong to a different period than the rare earths; however, because of their similarity in ionic radius, uranium and thorium may be incorporated in the structure of allanite. The radioactive elements accompany the rare earths in monazite, xenotime, bastnaesite, chevkinite, doverite, and keilhauite. Allanite has a lesser amount of radioactive elements captured in its structure than zircon, xenotime, or monazite, and it approximates the radioactivity of sphene, apatite, and the rare-earth carbonates.

Table 4 shows thorium to be high in the New Hampshire and Idaho minerals and low in the California minerals, regardless of rock type. Uranium is seen to be high in the allanites from granites regardless of the location.

Figures 1 and 2 show the relation of the calculated total percent eU to the birefringence and to the beta index of allanite samples.

Table 4.--Radioactive components of allanite from igneous rocks.

Rock and location	U 1/ (percent)	Th 1/ (percent)	Th/U	Th as percent eU	Calculated total eU (percent)	Indices of refraction α	β	γ	γ - α
New Hampshire									
Weathered Conway granite, Conway	0.0656	2.33	35.5	0.583	0.649	1.720	1.737	1.740	0.020
Fresh Conway granite, Conway	0.0540	0.92	15.1	0.202	0.256	1.721	1.738	1.742	0.021
Idaho									
Granodiorite, Stanley	0.0040	1.14	286.	0.286	0.296	1.761	1.776	1.780	0.019
Porphyritic granodiorite, Cascade	0.0036	1.05	291.	0.264	0.268	1.740	1.751	1.755	0.015
Granodiorite, Atlanta	0.0045	0.99	220.	0.249	0.253	1.752	1.768	1.771	0.019
California									
Rubidoux granite, fine, Riverside	0.0400	0.67	16.7	0.167	0.207	1.735	1.750	1.752	0.017
Stonewall granodiorite, Cuyamaca	0.0111	0.72	64.8	0.180	0.191	1.705	1.717	1.720	0.015
Woodson granodiorite, Temecula	0.0055	0.56	101.	0.141	0.146	1.735	1.750	1.753	0.018
Woodson granodiorite, Descanso	0.0102	0.38	37.2	0.097	0.107	1.745	1.760	1.763	0.018
Quartz monzonite, Basin Mountain	0.0078	0.35	44.6	0.088	0.096	1.750	1.766	1.770	0.020

1/ Analyst: A. M. Sherwood.

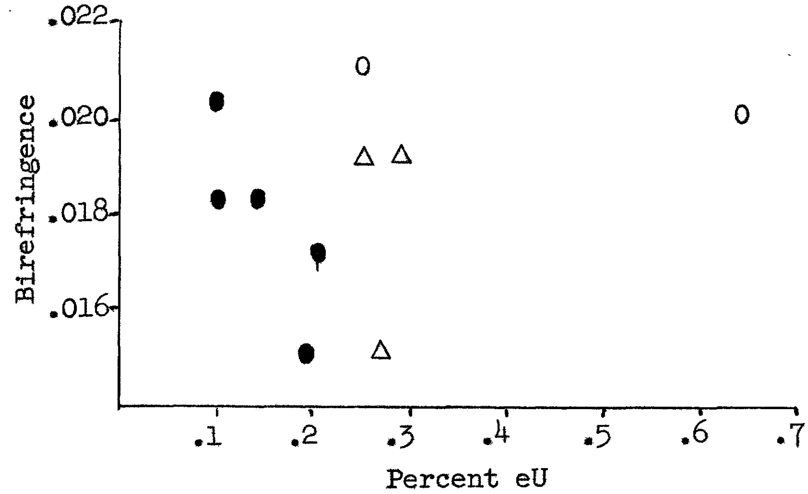

Figure 1.--Percent eU as related to birefringence in allanite from New Hampshire O, Idaho △, and California ●.

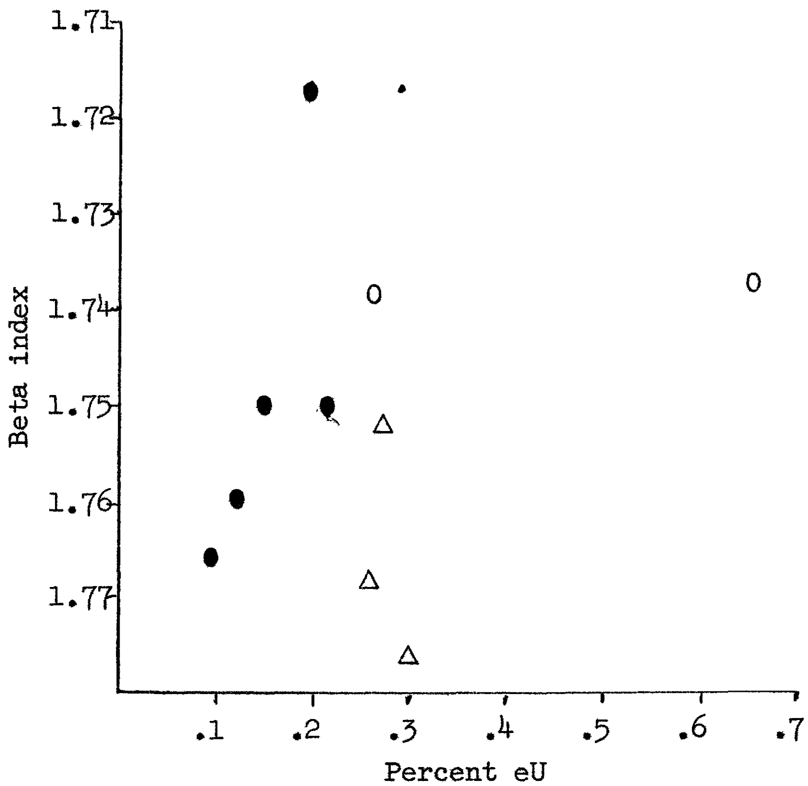

Figure 2.--Percent eU as related to beta index in allanite from New Hampshire O, Idaho △, and California ●.

A black high-index allanite occurring in late calcite-celestite veins cutting rare-earth-bearing carbonate rock from Mountain Pass, California (personal communication, H. W. Jaffe, 1955) has indices of refraction of α 1.790, β 1.812, and γ 1.818; a U content of 0.0018 percent and Th in the range of 0.01-0.1 percent. J. P. Marble (1940) describes isotropic allanite from the Barringer Hill, Texas, pegmatite with $\underline{n}$ = 1.716, as containing 0.715 percent Th and 0.033 percent U. Wells (1934) presents the analysis of allanite from a Wyoming pegmatite with 1.12 percent Th and 0.017 percent U. Hutton (1951) describes allanite from a pegmatite in Yosemite National Park, Calif., as containing 0.95 percent ThO_2, 0.015 percent UO_2, and optics α 1.769, β 1.782, and γ 1.791. Microscopic study of five specimens of black, vitreous allanite from pegmatites by E. S. Larsen, Jr., of the Geological Survey (Watson, 1917) describes a range from the isotropic to the birefracting forms of the mineral. The isotropic forms show indices of refraction from $\underline{n}$ = 1.60 to 1.72. In a study of the radioactivity of allanite from igneous rocks Hayase (1954) concluded that thorium was the major radioactive component, ranging from 0.5 to 1.6 percent.

The uranium content of rocks varies regionally, and generally in each region the more siliceous rocks have the higher uranium content. The relationship of high uranium allanite to high uranium rocks is apparent from observation (table 5), but it should be pointed out that only a percent or two of the total uranium in the rocks is traceable to allanite.

For comparison three samples of epidote were analyzed and show comparatively higher uranium contents for their rock type and area (table 6). Epidote is less abundant than allanite in the rocks studied. In all the rocks epidote was green and allanite was brown or black.

Table 5.--Uranium content of accessory allanite and of its host rocks. Analysts: A. M. Sherwood, M. Molloy, and M. Schnepfe.

Rock, location	U in allanite (ppm)	U in rock (ppm)
Granites		
Biotite granite, weathered, Conway, N. H.	656	12.0
Biotite granite, fresh, Conway, N. H.	540	13.0
Leucogranite, fine, Riverside, Calif.	400	3.7
Leucogranite, coarse, Riverside, Calif.	241	4.1
Albite granite, Sierra Nevada, Calif.	50	2.0
Granodiorites		
California, Mount Hole	208	5.9
Cuyamaca	111	1.5
Descanso	102	3.7
Temecula	55	1.2
Idaho, Atlanta	45	2.3
Stanley	40	0.8
Cascade	36	1.1
Quartz monzonites		
California, Pine Creek	91	7.7
Lamark	79	5.8
Basin Mountain	78	3.8
Quartz syenite		
Passaconaway, N. H.	39	4.6

Table 6.--Uranium content of epidote from California rocks.
Analyst: A. M. Sherwood

Rock	Uranium in epidote (ppm)
Mount Hole granodiorite	1310
Lamark quartz monzonite	220
Palisade quartz monzonite	180

PETROGRAPHY

Allanite was found to be present in the rocks studied in amounts ranging from 0.25 percent in the Cascade granodiorite to 0.005 percent in the Stonewall granodiorite. The abundance of allanite in a rock has no apparent direct relation to the uranium or thorium content of the rock or mineral and is not related to rock type or area. Of 81 rocks studied, 31 were found to contain allanite. No allanite was found in any of 10 siliceous lavas from the San Juan region of Colorado. None was found in 5 alkalic rocks from Sussex County, New Jersey. No allanite was found in any basalts, norites, gabbros, diorites, or nepheline rocks, although geologic literature describes occasional occurrences of allanite within such rock types (Iddings and Cross, 1885).

Table 7 lists the incidence and abundances of allanite in five suites of allanite-bearing rocks.

Table 7.--Incidence and abundance of accessory allanite.

Location	Number of rocks studied	Number of rocks with allanite	Percentage of allanite and rock type 0.X	0.0X	0.00X or less
Idaho batholith	14	6	1 grano-diorite	2 grano-diorites	3 grano-diorites
Sierra Nevada batholith, Calif.	6	4	--	3 quartz monzo-nites	1 granite
Southern Calif. batholith	34	13	1 granite	2 grano-diorites 1 granite	7 grano-diorites 1 quartz sye-nite 1 quartz mon-zonite
Sterling batholith, R. I.	4	3	--	1 grano-diorite	2 granites
White Mountain, N. H., batholith	8	5	1 granite	2 granites 1 quartz syenite	1 granite

METAMICTIZATION

Brögger (1893) in proposing the term metamict for certain rare-earth minerals suggested, "...The reason for the amorphous rearrangement of the molecules might perhaps be sought in the lesser stability which so complicated a crystal molecule as that of these minerals must have in the presence of outside influences."1/ He implied that the rare-earth minerals were so complex as to prevent them from being permanently combined in the crystalline state--a definition no longer accepted.

1/ Translated by A. Pabst (1952, p. 138).

Des Cloizeaux and Damour (1860) noted that isotropic allanite became birefringent upon heating and showed that both the isotropic and birefringent allanite existed both anhydrous and hydrated.

Goldschmidt and Thomassen (1924) describe the alteration of rare-earth minerals from the crystalline to the glassy state. The conclusion is reached that the important factor is the weak chemical bonding between rare earths and weak acids (silicic, tantalic). Goldschmidt proposed that in order for metamictization to take place the crystal lattice must have a weak enough ionic structure to permit decomposition and hydrolysis. Also, he proposed that it is necessary that radiation provide the energy to discharge the ions of the rare-earth elements. This radioactivity could be either from within or from without the crystal. Metamictization would thus occur as the ionic bond breaks by hydrolysis, and the lattice becomes isotropic.

Ellsworth (1925) states that "...all minerals containing UO_2 automatically oxidize themselves at a rate depending on the rate of uranium and thorium decomposition."

Hata (1939) in describing the weathering of allanite reports the altered portions as being conspicuously high in thoria and conspicuously low in the rare earths, as compared with the fresh part of the same mineral. As the result of leaching studies, Hata concluded that the alteration is likely to take place when the ratio of Fe_2O_3 to Al_2O_3 is less than 1.3 and the content of thoria is more than 1.5 percent. The other variables and the relative proportions of the rare earths were found to have no influence on the alteration.

Ueda and Korekawa (1954) suggest that the metamictization of allanite is due to the repeated expansion and quenching of the lattice, resulting in the formation of an aggregate of several phases in both the crystalline and amorphous state.

Allanite shows various degrees of metamictization and, in the extreme cases of some pegmatite specimens, approaches isotropism. The alteration of the allanite is suggested to be due to radiation from the uranium and thorium components which breaks the ionic bonds and permits the entry of water into the lattice of the mineral.

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