URANIUM AND THORIUM IN THE ACCESSORY ALLANITE OF IGNEOUS ROCKS[1]

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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 ranges from 0.005 to 0.25 per cent 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 the granites sampled, ranging from 0.004 to 0.066 per cent, whereas the thorium content is high or low regionally, ranging from 0.35 to 2.33 per cent. 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 depending 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 roll-type crusher which liberated the allanite cleanly. The allanite was generally found to concentrate in the 100- to 200-mesh size fraction. Allanite concentrates

* Rare earths.

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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 in a Frantz Isodynamic magnetic separator, allanite becoming magnetic between 0.4 and 0.6 amp. at cross and logitudinal settings of 10°. When epidote occurs with allanite, the separation becomes more difficult. The epidote floated and the allanite sank in methylene iodide saturated with iodoform (sp. gr. 3.45). Thus a minimum of hand-picking was required to obtain clean separates of the minerals.

Optical properties

All of the allanite studied was optically negative. The indices of refraction of the allanite were generally higher than those described in literature. This may be because the minerals generally described are the more metamict varieties from pegmatites. It is likely that the allanite described here is more abundant and more typical of the fresh mineral than the larger specimens obtained from pegmatites.

The optical data in Table 1 show that the α indices range from 1.690 to 1.775, β from 1.70 to 1.789, and γ from 1.706 to 1.791 with a possible error of ± 0.002. Allanite is reported to have indices as low as n 1.60

TABLE 1. INDICES OF REFRACTION OF ALLANITE FROM IGNEOUS ROCKS

Host rock	α	β	γ	Birefringence
Southern California batholith				
Coarse phase				
Granite from Rubidoux Mountain, Riverside	1.775	1.789	1.791	0.016
Fine Phase				
Granite from Rubidoux Mountain, Riverside	1.735	1.750	1.752	0.017
Woodson Mountain granodiorite, Descanso	1.745	1.760	1.763	0.018
Woodson Mountain granodiorite, Temecula	1.735	1.750	1.753	0.018
Woodson Mountain granodiorite, Rainbow	1.740	1.755	1.759	0.019
Woodson Mountain granodiorite, Elsinore	1.740	1.755	1.760	0.020
Granodiorite, Stonewall formation, Cuyamaca	1.705	1.717	1.720	0.015
Mount Hole granodiorite, Mount Hole	1.695	1.710	1.714	0.019
Tonalite, Aguanga	1.743	1.760	1.763	0.020
Sierra Nevada batholith				
Quartz monzonite, Basin Mountain	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 porphyritic quartz syenite, Passaconway	1.690	1.700	1.706	0.016

among its isotropic varieties. The birefringence, $\gamma-\alpha$, is shown to vary from 0.015 to 0.021 in the specimens studied. The indices of refraction of allanite from a single rock were found to be variable. The indices listed are representative. In the fresh Conway granite the allanite varies in α index from 1.695 to 1.739, and in the Cascade granodiorite the allanite varies in α index from 1.740 to 1.760.

There seems to be no clear relationship between the indices of refraction and the rock type. The Idaho minerals have consistently higher indices, whereas those from New Hampshire have lower indices. All of the allanite studied is from rocks of Late Jurassic or Cretaceous age (Larsen, 1948; Chapman, 1955; Hinds, 1934) with the exception of the New Hampshire samples which are of Mississippian age (Billings, 1945).

Chemistry

Chemical analysis

Chemical analyses (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 Per Cent, of Allanite from Cascade, Idaho; Conway, New Hampshire; and Riverside, California[1]

Constituent	Cascade granodiorite	Conway granite	Granite from Rubidoux Mountain, (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 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 batholith of New Hampshire, the Southern California batholith, the Sierra Nevada batholith, and the Idaho batholith. The rocks include fresh and weathered alkalic 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 thulium from the Basin Mountain mineral and the absence of lutetium 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 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 per cent, for K is 0.3 per cent, and for Li is 0.04 per cent. 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 Å to 4750 Å.

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 per cent U and one-quarter of the per cent Th. The minerals contain no detectable potassium.

TABLE 3. SEMIQUANTITATIVE SPECTROGRAPHIC ANALYSES OF ALLANITE
Analyst: Mona L. Franck

Rock	Fresh Conway granite	Weathered Conway granite	Granite from Rubidoux Mountain, fine phase	Woodson Mountain granodiorite	Woodson Mountain granodiorite	Granodiorite, Stonewall formation	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
Per Cent >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	

TABLE 4. RADIOACTIVE COMPONENTS OF ALLANITE FROM IGNEOUS ROCKS

Rock and location	U[1] (per cent)	Th[1] (per cent)	Th/U	Th as per cent eU	Calculated total eU (per cent)	Indices of refraction			$\gamma-\alpha$
						α	β	γ	
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									
Granite from Rubidoux Mountain, fine, Riverside	0.0400	0.67	16.7	0.167	0.207	1.735	1.750	1.752	0.017
Granodiorite, Stonewall formation, Cuyamaca	0.0111	0.72	64.8	0.180	0.191	1.705	1.717	1.720	0.015
Woodson Mountain granodiorite, Temecula	0.0055	0.56	101.	0.141	0.146	1.735	1.750	1.753	0.018
Woodson Mountain 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.

Partly because of their similarity to the rare earths in ionic radius, uranium and thorium may be incorporated in the structure of allanite. The radioactive elements accompany the rare earths in many minerals, for example, in monazite, xenotime, bastnaesite, chevkinite, doverite, and keilhauite. In the rocks studied allanite has a lesser amount of radioactive elements captured in its structure than ziron, xenotime, or monazite, and it generally approximates the radioactivity of sphene, apatite, and the rare-earth carbonates.

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 rocks (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
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		
Mt. Tom quadrangle	91	7.7
Mt. Goddard quadrangle	79	5.8
Mt. Goddard quadrangle	78	3.8
Big Pine quadrangle	50	2.0
Quartz syenite		
Passaconway, N. H.	39	4.6

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 from both localities listed in Table 5.

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

A black high-index allanite occurring in late calcite-celestite veins

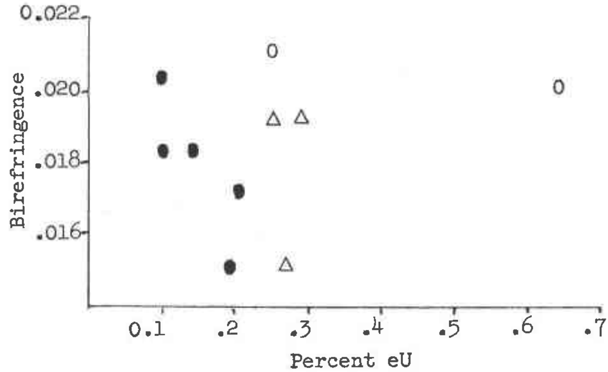

FIG. 1. Per cent eU as related to birefringence in allanite from New Hampshire ○, Idaho △, and California ●.

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 per cent and Th in the range of 0.01–0.1 per cent. J. P. Marble (1940) described isotropic allanite from the Baringer Hill, Texas, pegmatite with $n=1.716$, as containing 0.715 per cent Th and 0.033 per cent U. Wells (1934) pre-

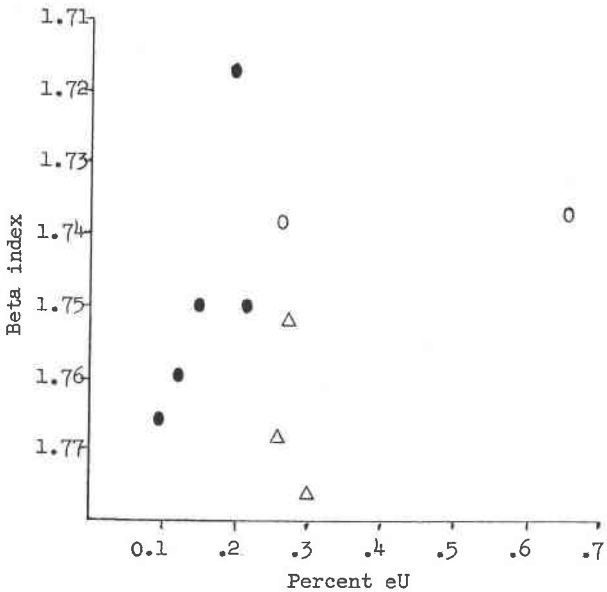

FIG. 2. Per cent eU as related to beta index in allanite from New Hampshire ○, Idaho △, and California ●.

sented the analysis of allanite from a Wyoming pegmatite with 1.12 per cent Th and 0.017 per cent U. Hutton (1951) describes allanite from a pegmatite in Yosemite National Park, Calif., as containing 0.95 per cent ThO_2, 0.015 per cent UO_2, and optics α 1.769, β 1.782, and γ 1.791. In a microscopic study of five specimens of black, vitreous allanite from pegmatites by E. S. Larsen, Jr., of the Geological Survey (Watson, 1917) a range from the isotropic to the birefracting forms of the mineral is

TABLE 6. URANIUM CONTENT OF EPIDOTE FROM CALIFORNIA ROCKS
Analyst: A. M. Sherwood

Rock	Uranium in epidote (ppm)
Mount Hole granodiorite	1310
Quartz monzonite, Mt. Goddard quadrangle	220
Quartz monzonite, Big Pine quadrangle	180

described. The isotropic forms show indices of refraction from $n = 1.60$ 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 per cent.

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 per cent 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.

PETROGRAPHY

Allanite was found to be present in the rocks studied in amounts ranging from 0.25 per cent in the granodiorite from Cascade to 0.005 per cent in the granodiorite from the Stonewall formation. 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 five 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.

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

Table 7. Incidence and Abundance of Accessory Allanite

Location	Number of 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 granodiorite	2 granodiorites	3 granodiorites
Sierra Nevada batholith, Calif.	6	4	—	3 quartz monzonites	1 granite
Southern California batholith	34	13	1 granite	2 granodiorites 1 granite	7 granodiorites 1 quartz syenite 1 quartz monzonite
Sterling batholith, R. I.	4	3	—	1 granodiorite	2 granites
White Mountain batholith, N. H.	8	5	1 granite	2 granites 1 quartz syenite	1 granite

complicated a crystal molecule as that of these minerals must have in the presence of outside influences."* 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.

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

Goldschmidt and Thomassen (1924) described the alteration of rare-earth minerals from the crystalline to the glassy state. They concluded that the important factor is the weak chemical bonding between rare earths and weak acids (silicic, tantalic). Goldschmidt proposed that 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

* Translated by A. Pabst (1952, p. 138).

thus occur as the ionic bond breaks by hydrolysis, and the lattice becomes isotropic.

Ellsworth (1925) stated 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 per cent. 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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