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ALLANITE FROM WADI EL GEMAL AREA, EASTERN DESERT OF EGYPT, AND ITS RADIOACTIVITY

Amin R. Gindy, *Department of Geology, Alexandria University, Alexandria, Egypt, U.A.R.*

Introduction

The Geological Survey of Egypt recently reported small pegmatite dikes and veins carrying sporadic large crystals of allanite in the Precambrian basement complex rocks of Wadi El Gemal area, near the Red Sea coast, Eastern Desert of Egypt. Sadek (1953) indicated the localities of these occurrences and gave a brief report of other mineral deposits there. The allanite-bearing pegmatites are particularly frequent in the rectangle limited by longitudes 34° 40′–34° 50′ and latitudes 24° 30′–24° 40′ where they seem to be closely associated with a complex of xenolithic and gneissic tonalites, granodiorites and diorites. These pegmatite dikes and veins are of rather simple mineralogy and are mainly made up of unzoned acid oligoclase with smaller amounts of quartz and potash-rich feldspars. Allanite is almost the only colored constituent present; other accessories being either absent or in rare tiny grains. These include opaque ores, apatite, sphene and late hydrothermal clinozoisite, epidote and muscovite. The allanite-pegmatites show signs of late stage hydrothermal alterations; plagioclase being variably sericitized or kaolinized and allanite is also affected as described below.

Allanite

Allanite occurs erratically in the pegmatite bodies, sometimes in large euhedral to subhedral crystals measuring up to 3 inches in length (presumably parallel to b) and have a cross section of about $1\frac{1}{2}$ square inches. Usually it occurs in stubby subhedral crystals about a square inch in cross section. Much smaller crystals are also present in the feldspar-quartz matrix.

The mineral is opaque with a brownish black color and resinous luster on fresh broken surfaces. Along major fractures or along its contact with the matrix of the rock, the mineral has a somewhat rusty surface. Thin splinters of the mineral are translucent in dark or smoky amber colors. The powdered mineral has a dark buff grayish color and a greasy luster. Surface striations and cleavages parallel to b, the length of the crystal, are sometimes developed in the large crystals. Specific gravity ranges from 3.32 to 3.75. Hardness 5.5. The mineral in small fragments or powder readily swells and intumesces when heated in a platinum loop. The magnetism of the dark brown fused mass is variable, probably indicating different contents of iron. A borax bead gives a test for iron. Boiling HCl briskly decomposes the mineral, leaving a white but not gelatinous precipitate (cf. Hutton, 1950, p. 244–7).

The mineral apparently varies widely in composition in the different samples studied. In individual large crystals, however, zonal growths of different or contrasting compositions are generally slight or entirely lacking. When present, they are indicated by slight and patchy differences in color or birefringence. These differences in composition within a large crystal are revealed better when the mineral is altered. Fresh, birefringent crystals are pleochroic with $Z \simeq Y$ olive green and X pale yellowish green or straw yellow. The darker fresh varieties are more pleochroic and also richer in iron and radioelements than fresh allanites of paler pleochroic colors. In the fresh allanites, n varies between 1.74 and 1.70. 2V is negative and ranges from 77° to 84°.

In thin section, the large cyrstals appear to have crystallized early. Sometimes their outer borders show mild sinuous embayments as if they had been partly resorbed at a late stage in their history. The resorbed margins were later framed by a fringe of complicated fine granular aggregates of clinozoisite, epidote, red or orange allanite, magnetite or hematite, some quartz or even albite (Fig. 1). Near their outer borders, fresh large subhedral allanite plates may show some signs of irregularity in original composition and patches of colorless clinozoisite appear. Relatively late hydrothermal clinozoisite commonly traverses the broad allanite plates in tiny veinlets along pre-existing fractures. These veinlets do not usually extend beyond the allanite plate into the quartz-feldspar
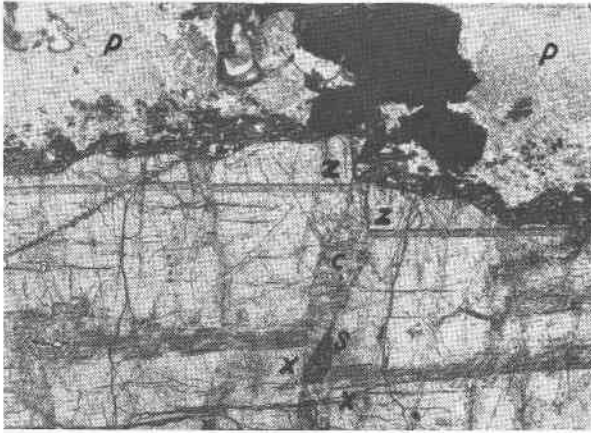
FIG. 1. Photomicrograph of part of the outer border of a large allanite crystal showing a resorbed sinuous border outlined by a dark fine-granular intergrowth of yellow to red allanite, clinozoisite and opaque grains. A clinozoisite vein (*c*) with a sphene crystal (*s*) traverses the allanite plate along a micro-fault indicated by the displacement of two sub-parallel lightly altered zones (*z* and *x*) within the allanite plate. This particular allanite plate is fairly fresh and anisotropic. It is also the least radioactive variety encountered. Matrix of altered acid oligoclase (*p*) with opaque grains. Ordinary light, ×22.

matrix, but at the outer boundaries of allanite they merge with the late peripheral clinozoisite growths. The clinozoisite veinlets may also contain rare crystals of sphene, also apatite and quartz.

ALTERATION OF ALLANITE

All allanites from Wadi El Gemal show replacement by a bright yellowish, orange, red or brown variety around their borders and along some internal cleavages and fractures; rarely in certain internal zones that seem to be of a slightly different composition more susceptible to alteration. In a more advanced stage of alteration, clouds of tiny hematite-like lamellae are formed. In the iron-rich allanites, these lamellae become concentrated into tiny brown to black opaque irregular clots spangling the altered mineral. The small crystals of allanite in the quartz-feldspar matrix of the pegmatites are always entirely replaced by this deeply colored allanite. In the larger crystals of originally weak radio-activity or in isotropized ones of higher radioactivity some internal parts of the plates remain clear, and retain the original pale olive green color.

The apparently fresh and homogeneous islands remaining in the relatively iron-rich strongly altered allanite crystals, while fairly clear and retaining their original olive green color, become almost non-pleochroic and non-birefringent. Under crossed nicols, this isotropism is not perfect

or homogeneous but is rather mottled throughout by slightly birefringent small patches which, however, give no interference figures. Further alteration of this clear isotropic allanite gives very complex and intimate fine granular intergrowths of the reddish birefringent allanite and a low birefrigent white or gray mineral (clinozoisite?) accompanied by a release of hematite-like lamellae, or, in intensified alterations, by the dark brown, almost opaque (amorphous?) ferruginous clots and veinlets. Several granules of this secondary reddish allanite may retain a common optical continuity over some patches of this complex intergrowth. In other parts, pleochroic greenish epidote and colorless clinozoisite are among the alteration products. Indeed some of the relatively wider and better defined clinozoisite veinlets traversing the allanite may taper near their narrowing ends deep inside the host into epidote-veinlets or epidote-clinozoisite veinlets that soon merge and lose their identity among the fine granular alteration products of the host. Elsewhere in such allanite crystals, relatively large epidote and clinozoisite patches in the altered parts seem to be of a straightforward replacement origin. No isotropized allanite was observed in the transitional stages of alteration of the originally weakly radioactive and usually iron-poor allanite specimens. Also no definite green epidote is found among their alteration products.

As the clear relatively more radioactive and iron-rich allanites are isotropized and are also much more strongly altered and replaced by the deeply colored allanite-hematite-clinozoisite-epidote aggregates than the least radioactive and iron-poor allanites which are never isotropized but undergo a milder alteration to the reddish allanite-hematite or red allanite-clinozoisite-aggregates, it seems convenient to distinguish between two somewhat independent lines of changes; (*a*) isotropization, and (*b*) replacement by the deeply colored allanite-hematite-clinozoisite aggregates. Isotropization may be tentatively ascribed to metamictization by radiation with hydrothermal alterations taking a doubtful part. Hydrothermal alteration is, however, directly responsible for the deep red allanite-hematite-clinozoisite aggregates irrespective of the degree of radioactivity of the original allanite. Nevertheless, the original composition of the allanite seemed to present an important measure of its susceptibility to the hydrothermal alterations, the more ferruginous and more radioactive allanites being much more affected. Damage and breakdown of ionic bondages brought about by radiation would certainly help the hydrothermal alteration. Completely oxidized and hydrolysed outer parts of the originally strongly radioactive crystals are transformed into deep brown opaque pseudomorphs that are perhaps amorphous. In the Elberton granites of Georgia, Silver and Grunenfelder (1957) have considered the principal alteration of accessory allanite to have resulted not

from simple metamictization but rather from reactive deuteric or later solutions.

Radioactivity of Allanite

Radioactivity of allanite was studied from polished specimens and uncovered thin sections previously stored in a dry container for at least two months in order to restore original radioactive equilibrium. Ilford Nuclear Research plates, emulsion types G.5. and C.2. were used. An exposure of two to three weeks of specimens in close contact with G.5. plates was sufficient to give good megascopic autoradiographs (Fig. 2, B). Similar autoradiographs were also obtained on medical double-coated x-ray sheets. For quantitative work on alpha tracks, Ilford C.2. plates were used at varying exposures from 5 to 20 days and special precautions were taken to guard against fading of latent images in the humid atmosphere of Alexandria.

The total surface radioactivity of the large clear allanite crystals studied in the pegmatite specimens ranged from 0.087 to 0.672 $\alpha/cm.^2$ /sec. (eU_3O_8 content of about 0.066 to 0.51%) but large clear individual crystals in the same specimen do not usually differ widely in their radioactivity. The most frequent range of activity for fresh crystals is between 0.35 and 0.5 $\alpha/cm.^2/sec.$ Clear crystals of weaker radioactivity (with a surface activity perhaps up to about 0.4 $\alpha/cm.^2/sec.$) were mostly birefringent and looked fairly homogeneous in thin section as well as in the spatial distribution of alpha tracks in their corresponding autoradiographs. Crystals of higher radioactivity generally showed a marked alteration and a corresponding irregular pattern in their autoradiographs (Fig. 2, A and B). The highest values of radioactivity are measured from the relatively clear isotropized and least affected relict areas within the altered crystals. In such areas, radioactivity is still much more evenly distributed than in the surrounding altered areas. In the altered parts of the allanite crystals a clear decrease in radioactivity indicates leaching away of part of the radioelements together with iron and probably other cations. Radioactivity of the pleochroic yellowish or reddish alteration variety of allanite is very variable but may equal that of the parent mineral. In the altered parts, however, some highly radioactive spots are occasionally present. These are not produced by highly active mineral inclusions but are probably remains from old radiocolloid aggregates as mentioned later.

The total surface area of the thin section of allanite shown in Fig. 2, A (excluding areas occupied by the large clinozoisite veinlets) has a total activity of 0.4 $\alpha/cm.^2/sec.$ In the clear isotropized core, the activity is 0.59 while in many altered parts it is down to 0.23. The clear clinozoisite
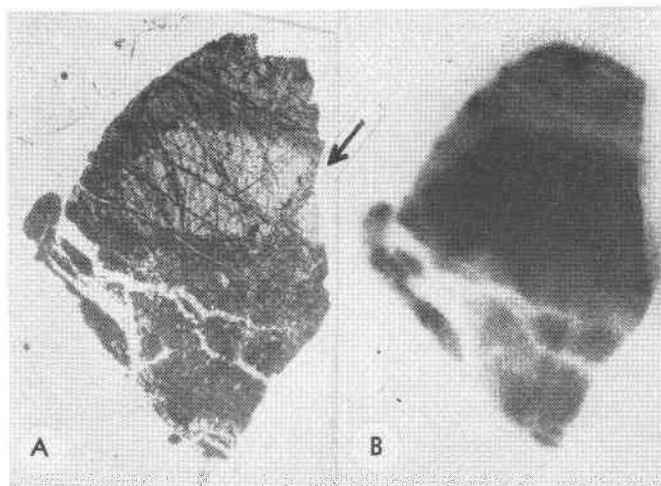
Fig. 2. (A) Thin section of part of a large allanite crystal showing a rather clear less altered isotropized island surrounded by a darker much altered area with clinozoisite veinlets × about $2\frac{1}{2}$. Arrow points to a narrow alteration channel in the less altered more radioactive island. (B) Autoradiograph of the same specimen in (A) on Ilford G.5. plate. Notice the higher radioactivity of the less altered island compared with the altered part and the non-radioactivity of the clinozoisite veinlets. The autoradiograph was originally made from the polished surface of a thick slice of allanite which was later mounted on a slide and thinned from the other surface to standard thickness. A part of the upper edge of specimen in (A) was lost in the process.

veinlets are found to be almost completely non-radioactive (Fig. 2, B). The few alpha tracks emitted from them were actually traced to ferruginous dark brown particles, and films that had been leached from the adjacent much altered parts of the host. The photograph in Fig. 2 may give the impression of an originally zoned allanite crystal with an internal core of a different composition and a higher radioactivity than the outer, now turbid and altered zone. This is disproved by the irregular distribution of radioactivity in the entire crystal and by the fact that narrow zones of alteration identical to those of the outer zones do cross at random directions into the clear core, producing narrow zones of reduced activity. These narrow zones do not show well in the sliced fragment under the small magnification used. Fragments from another part of the same allanite crystal, part of which is shown in Fig. 2, A, were powdered and part of the powder was boiled in dilute HCl (1:5). The powder and the white pulverulent material resulting from the acid leaching had specific activities of 0.44 and 0.0025 α/cm.^{2}/sec. respectively.

In order to estimate roughly the ThO_2 and U_3O_8 contents, measurement of lengths of individual alpha tracks was attempted on autoradiographs of two clear allanites giving a fairly uniform pattern of alpha

track population. The two allanite slices had specific activities of 0.25 and 0.35 and the corresponding Th/U ratios were found to be 2.04 and 1.748 respectively. This would indicate roughly a content of about 0.225% ThO_2 and 0.110% U_3O_8 for the first slice, and of 0.257% ThO_2 and 0.147 U_3O_8 for the second slice. If the average Th/U ratio of 1.9 is considered as fairly constant for all the fresh or clear allanites, then the highest thorium and uranium contents of the examined specimens (corresponding to an activity of 0.672) would be about 0.578 and 0.304% respectively. A. I. Ghozlan (personal communication, 1958) determined the Th/U ratios for three large allanite slices from Wadi El Gemal area by the same technique and gave the following values 1.785, 1.685 and 1.485. He further checked the last two values by the second technique available in such studies, *viz.* by counting the relative frequency of the various alpha "stars" produced by the successive disintegration of the short-lived decay products (Bremner, 1951). He obtained Th/U ratios of 1.787 and 1.805 respectively. If all the preceding Th/U values were correct, then the present fresh allanites would be rather unusual in containing a much higher amount of uranium relative to thorium than is usually given in the literature on allanite. It is not quite certain at present whether the five allanite slices on which Th/U ratios were determined all come from the same pegmatite dike or from different pegmatite bodies. Th/U ratios and radioelement contents of the altered allanites were not studied in detail but there are indications of the well known higher release and mobility of uranium relative to thorium during hydrothermal alterations and weathering of allanite. It is therefore advisable, when Th/U ratios for allanites are given to mention the degree of alteration of the minerals.

Radiocolloid aggregates and zones of higher radioactivity in altered allanites

Tiny highly radioactive spots in autoradiographs of allanite were always traced to the strongly altered and leached parts (Fig. 3) and were not emitted by areas of either the fresh anisotropic allanite or the clear isotropized allanite. It may be recalled that it is in such highly altered parts that the radioactivity is strongly reduced. Fig. 3, A, shows the most radioactive spot so far observed in the altered allanite. Judging from the boundary line enclosing all vertical and nearly vertical alpha tracks of this spray in the autoradiographs, the source must have been an area below 14 square microns with an activity of over 6000 $\alpha/cm.^2/sec.$ This active spray was traced to a spot in an opaque dark brown (amorphous?) ferruginous veinlet leached in a channel within the altered allanite. In one odd example of a strongly altered outer border of an allanite crystal, these active spots occurred at a frequency of about 340 sprays per square centimeter of the surface area of the altered mineral, but usually they are much fewer. Slight repolishing of the allanite surface

causes earlier sprays to disappear but new ones may appear elsewhere in the corresponding autoradiographs. Investigation of the lengths of alpha tracks emitted by such sprays in autoradiographs of suitable periods of exposure indicated that these tracks do not exceed in length those emitted by RaC′, the longest tracks of the uranium family. Such sprays are thus most probably produced by segregation, during leaching of the altered allanite, of daughter products of the uranium series, and may represent radiocolloid aggregates (cf. Yagoda, 1946).
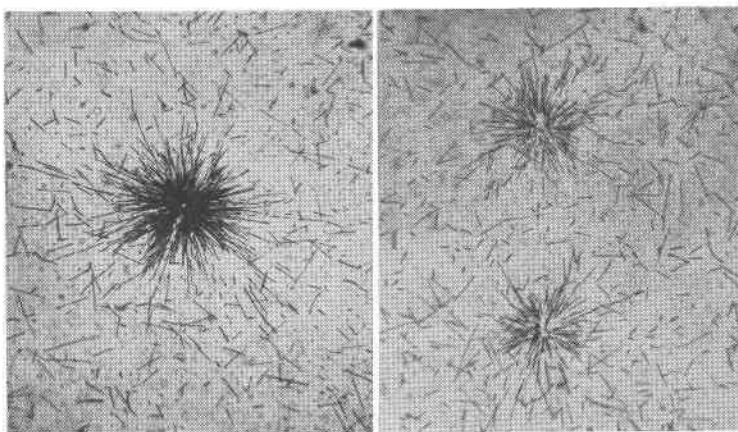

FIG. 3. (A) (left) Photomicrograph of an alpha autoradiograph of an altered allanite, showing a strong spray of alpha tracks emitted by a center probably suggesting an original radiocolloid aggregate, ×200. (B) (right) Two weaker sprays from the same autoradiograph of (A), probably representing older segregates than the one shown in A, ×200.

Beside these highly active sprays, the autoradiographs also show a few very narrow ribbon-like, oval or ill defined small zones where alpha tracks show some marked concentration. These zones were also located in the altered parts of allanite, particularly at the strongly altered allanite margins opposite the veinlets of clinozoisite, clinozoisite-quartz or clino-zoisite-epidote-quartz. These zones are mostly traced to areas occupied by the dark brown, almost strongly altered variety of allanite or its opaque pseudomorphs and alteration products

The radioactivity of all such narrow zones was not observed to exceed $3\alpha/cm.^2/sec.$ and may be about seven times higher than the radioactivity of the remaining fresh parts of the corresponding allanite and more than ten times higher than the average radioactivity of the ordinary altered parts. It may be tempting to consider them as relics after old incipient stages in radiocolloid development. Long ThC′ tracks are present in them but these can be accounted for by the background radiation of thorium in

the altered allanite. They may be also due to concentration of the radio-elements in some of the new red-brown varieties of allanite after the breakdown of the original allanite to red brown allanite, clinozoisite and epidote. Compared with clinozoisite, green epidote granules occasionally found in the clinozoisite veinlets are slightly more radioactive.

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A DIFFRACTOMETER MOUNT FOR SMALL SAMPLES*

A. J. GUDE, 3rd AND JOHN C. HATHAWAY,
U. S. Geological Survey, Denver 25, Colorado

Buerger and Kennedy (1958) discuss the problem of scattered x-radiation from a diffractometer sample holder and describe improved results using a BT-cut quartz oscillator plate. They point out that when a small amount of sample is supported in the x-ray path, background noise from various mounting media yields random deflections that may be indistinguishable from low-intensity peaks of the diffraction pattern. Specimens large enough to be packed, pelleted, or slabbed are quasi-infinite in thickness and thus mask the background contributed by the holder or support. However, where samples are very small, any contribution from the substrate may add significantly to the background scattering.

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