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RADIOACTIVE HALOS[1]

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(d) m.(Bi77 T85 Tf 0.46) Tj 3 0 TD /F1 9.750 Tw

Th halo.

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Schet5 TD /F-0.03975 Tf 0.0271 1 Tf 0.0271 Tc 0d 975 Tf 0.4605 Tc 0 Tw ((d 1852 Tj 36 0 TD 0 0 0009w

Figure 1 The scale for the photomicrographs is 1 cm equivalent to 45 mm. (a) Schematic drawing of ^{238}U halo with radii proportional to a-ranges in air. *(b)* Schematic drawing of ^{232}Th halo. *(c)* ^{235}U halo in biotite formed by sequential a-decay of the ^{238}U decay series. *(d)* ^{232}

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Quantitative evidence results from using newer methods that permit observation of single fission tracks in biotite (Fleischer, Price & Walker 5*)*. After appropriate etching, an embryonic U halo (only first ring visible) exhibits a cluster of 20 to 30 ^{238}U

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previously associated with the halo rings became almost nonexistent. Clearly it is not possible to obtain reliable standards for radii comparison using this procedure.

Further attempts to compare halo radii with equivalent mineral ranges derived from a-ranges in air (20) are also unsatisfactory because of uncertainties in the air-mineral conversion factor. Comparison of halo radii with a-ranges calculated from the Bragg-Kleeman (or a similar) relation is possible, but necessarily assumes that ring coloration extends the full a-range. Now newer techniques do allow appropriate standards to be developed, but before discussing these we first digress to examine in more detail halos in different growth stages.

Radiohalos in fluorite will be used as examples. Figure l*e* shows an embryonic U halo wherein only the first two rings are prominent. Figure 1*f* shows the normal or intermediate stage U halo wherein all rings may be detected. The U halo in Figure l*g* is overexposed in the inner halo region, resulting in coloration reversal and the formation of a diminutive ghost ring in the center. Figure l*h* shows two other partially reversed U halos, one of which shows the diminutive inner ring, while the other has experienced complete obliteration of all the inner rings. The U halo in Figure 1*i* is even more overexposed, and encroaching reversal effects have given rise to another ghost ring just inside the outermost periphery. Figure l*j* shows a still more highly overexposed U halo in which second-stage reversal effects have produced spurious ghost rings that are unrelated to the terminal a–ranges.

Tracing out the above pattern of U halo development in fluorite is no straightforward task. Only by observing differential growth increments in thousands of halos (produced of course by differential amounts of U in the halo inclusions) is it possible to construct the sequence shown. Earlier investigators (12, 23) as well as a later one (Gentry 28, 30) at one time erred in inventing new a–activity to account for some of the aforementioned ghost rings. Clearly a one-to-one correspondence between halo radius and a–energy is not always valid.

We return to the discussion of the constancy of a–decay energies and the problem of developing reliable standards for halo radii comparisons. The most direct technique is to irradiate halo-containing minerals with a sufficient dose of monoenergetic ^{4}He ions until halo-like coloration develops (Gentry 8, 24). If reciprocity holds, the ^{4}He ions will produce a coloration band (see Figure 1*k)* that in theory is equivalent in depth or size to a halo radius produced by the same energy a–particles. These induced coloration band (CB) sizes then form the standards against which halo radii may be compared. [Interestingly a densitometer profile of the CB in Figure 1*k* shows a marked resemblance to the shape of the Bragg ionization curve, thus providing some basis for the superposition procedure used by Henderson (20) and Joly (15).] However, the actual comparison procedure requires that certain additional factors be considered.

First, in some minerals, especially biotite, this reviewer has found that halo radii are somewhat dose dependent; darker rings show slightly higher radii than faint rings (24). (CB sizes show a similar effect, implying coloration intensities in natural and induced specimens should be matched before size comparisons are made.) This dose effect is often masked by subtle halo radius variations produced by attenuation of the as emitted within the inclusion itself. For example, when the inclusion (e.g. zircon) is more dense than the host mineral (e.g. biotite), slightly smaller radii result in embryonic halos; extreme values are reached only after a heavy dose. However, a heavy dose means a dark halo which tends to obscure the inner halo rings, making measurements difficult.

Although the finite inclusion size renders all radii measurements uncertain to a degree, there are two cases when this correction is minimized. For *densely colored* halos surrounding large inclusions (e.g. see Figure 2*c*), the only reasonable radius measurements are those made from the inclusion edge. In the other case for halos in biotite with only tiny 1-mm inclusions, this reviewer arbitrarily makes no correction for inclusion size.

In fluorite the situation is different. Here the effect of finite inclusion size is minimized because some halos exist with nuclei only 0.5 pm or less in diameter. Yet in contrast to halos in biotite, there is evidence that in some cases radioactivity is surficially distributed on the inclusion, implying halo radii be measured from the inclusion edge. Fortunately, the halo ring size in this mineral is only very minimally dependent on a-dose so that even embryonic halo rings form at practically maximum size (22—24).

Table I shows U halo radii measurements in biotite, fluorite, and cordierite (19, 20, 23—
by accelerator ^{4}He ion beams of varying energy in the same minerals (Gentry 24). Note that the comparison between fluorite CB sizes and halo radii is quite good, while in biotite the halo radii are somewhat smaller than the equivalent CB sizes. As discussed earlier, this deviation in biotite is likely due to the effects of a finite inclusion size plus the dose-dependent radius effect rather than a change in a–energy.[2] Note also the comparison between CB sizes and halo radii in cordierite. Here the agreement is fair even though the U halos in cordierite possessed large inclusions, necessitating that halo radii be measured from the inclusion edge (Mahadevan 0.10.

Interestingly densitometer profiles of some of my U halos show the same small halo rings that Henderson (2010tnoted, but these appear to be only artifacts of the coloration process and are not included in Table 1.

With respect to the decay rate question, Spector (3110thas argued that the differences between Henderson et al (2010thalo radii measurements and equivalent air mineral ranges present a case for a variable 2. In the light of the above experimental uncertainties, this conclusion is not necessarily valid. On the other hand, Gentry (24)thas shown that even exact agreement between halo radii and corresponding CB sizes does not necessarily imply an invariant A and in fact uncertainties in radius measurements alone preclude establishing the stability of **it** for ^{238}U to more than 35%.

[2] My U halo radii in mica show in the G colum of Table 1 give the range of radii sizes in this particular mica.

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Table 1 Halo radii and induced coloration band (CB) size measurements [a]

^{4}He Induced Coloration Band Sizes (㎜)					E (MeV)	Nuclide	U Halo Raddi (㎜)						Po Halo Radii (㎜) in Mica						Po Halo Radii (㎜) in Fluorite	
Mica			Fluo-rite	Cordi-erite			Mica			Fluorite		Cordi-erite	^{210}Po		^{214}Po		^{218}Po		^{210}Po	^{218}Po
G_L	G_M	G_D	G	G			K-L	H	G	S	G	M	H	$G_{L?\ D}$	H	G_L	H	$G_{L?\ M}$	G	G

? ? TD 0.25 0 /45 0.75 re f 567 639 0.Tw ( ) Tj 18 0 TD 0.087 Tc 0 Tw (?) Tj 4.5 0 TD 0 Tc 0.1874 459.75 606 0.75 10.5 re f 481.5 606 0.75 10.5 re f 502.5 6

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Po type halos along conduits in biotite. Those Po halos he found occurring apart from conduits (similar to those found by Gentry in Figure 1*p, r, s)* were more difficult to account for. Here a very qualitative laminar flow hypothesis was proposed. The intervention of World War II and his untimely demise soon thereafter prevented Henderson from determining whether these explanations were valid. [Although Henderson (32) suggested that such types existed, the halo in Figure 1*u* was discovered only recently by Gentry and cannot be explained on the basis of U daughter a -activity alone.]

Now the reason for the various attempts to account for Po halos by some sort of secondary process is quite simple; the half-lives of the respective Po isotopes are far too short to be reconciled with slow magmatic cooling rates for Po-bearing rocks such as granites ($T_{½}$ = 3 min for ^{218}Po).

Yet Gentry (6), by using fission-track and a-recoil techniques, found no evidence for a secondary origin of those Po halos in biotite, which occurred apart from conduits (cf Figure 1*p, r, s).* Consistent with the ring structure, fission-track analysis of the Po halo inclusions showed very little, if any, U. Further, the a-recoil technique (Huang & Walker 33), which permits the observation of a single a-recoil pit in biotite, was employed to measure the distribution of decayed a-radioactivity in regions both adjacent to and far removed from Po halo inclusions. No differences in a-recoil density were noted in the two areas. If U daughter a-activity had fed the Po inclusions, a significantly higher a-recoil density would have been in evidence.

One solution to this dilemma is the suggestion (24) that the parent nuclides of the Po in the halo inclusions were long half-life *b*-decaying isomers that may yet exist. Halo ring structure allows this, for *b*-radioation produces no coloration in halos in biotite, fluorite, or cordierite. [Laemmlein (34) reported *b*-halos in a certain quartz sample, but this is unconfirmed.] If such isomers existed, Po, U, and Th halos could all be explained by assuming that minute quantities of the respective nuclides were incorporated into separate halo inclusions either before or coincident with the host rock crystallization.

Dwarf Halos

Joly (15) reported the existence of dwarf halos with radii — ~*5.2* and *8.5mm* in the black micas from the pegmatite quarry at Ytterby near Stockholm. A rather unsatisfactory attempt has been made (7) to identify the 5.2-mm dwarf halo with ^{147}Sm a-decay (E_a = 2.24 MeV). Dwarf halos are extremely rare and erratically distributed even in the few mica samples which contain them. Joly (15) considered their radioactive origin beyond question and attributed their bleached appearance to radiation overexposure or to elevated temperatures.

Gentry (28) has also found dwarf halos in a mica specimen from Ytterby, and the sizes are difficult to correlate with a-decay systematics of known radioactive nuclides. The smallest dwarf halos range from only 1.5 to about 2.5 mm with associated a-energies in the 1MeV range (Figure 2a). The half-lives of known radionuclides are in excess of 1013 years for a-decay energies of 2 MeV or less. Such weakly active nuclides almost escape detection and would hardly be expected to produce a halo. Other dwarf halos with radii from 3 to 11 mm (Figure *2b)* correspond to a-energies of ~1.1 to 3.4 MeV. Some of these dwarf halos reflect coloration differences possibly due to varying amounts of parent radionuclides in the inclusions. In this respect the occurrence of dual-ring dwarf halos also suggests a radioactive origin. Although Gentry has not yet seen dwarf halos in cordierite, Mahadevan's (25) report of equivalent dwarf sizes in this mineral lends much credence to the radioactive origin of such halos. Whether there exists a causal relation between the dwarf halos and the previously reported, though unconfirmed, low-energy a-activity found by Bruki, Hernegger & Hilbert *(35)* is presently open to question. As noted earlier, the dwarf rings which Henderson (20) reported in some U halos appear to be only artifacts of the coloration process.

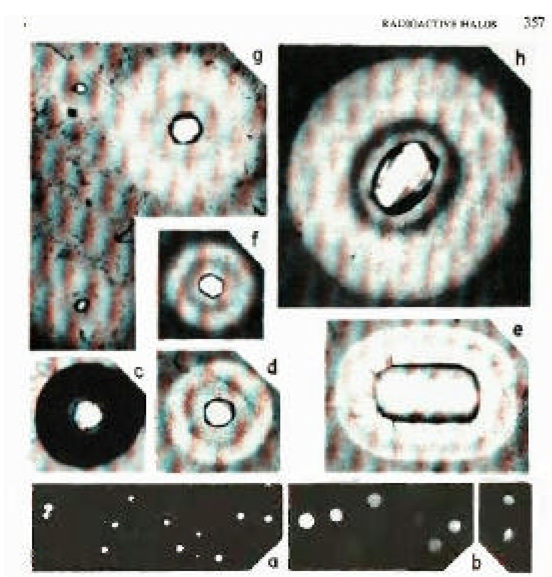

Figure 2 The scale for all photographs is 1 cm = 50 mm. *(a)* Dwarf halos (? 2-mm radius) in Ytterby mica. *(b)* Dwarf halos (3 mm? *r* ? 9 mm) in Ytterby mica. *(c)* Overexposed Th halo in ordinary biotite. *(d)* Th halo in Madagascar mica. *(e)* Th halo in Madagascar mica with a larger inclusion. *(f)* U halo in Madagascar mica. (g) Giant halo of ? 65-mm radius, and two light Th halos (Madagascar mica). *(h)* Giant halo of ? 90-mm radius, Madagascar mica.

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X Halos and Other Intermediate Size Varieties

Still rarer than the dwarf halos are the X halos first reported by Joly (15) in the micas from Ytterby and Arendal (Norway). Later van der Lingen (16) reported halos of similar dimensions occurred in a granite near Capetown. According to Joly (15) the inside ring of the X halo may be somewhat diffuse and measures about 8.5 to 9.8 mm in radius, corresponding to an E_a of about 2.9 to 3.2 MeV. The bleached rings extend out to a radius of about 14 to 15 mm, and sometimes an adjacent dark ring is evident at about 17 mm (E_a = 4.4 to 5.0 MeV). The outer wide band extends to approximately 28 mm corresponding to an E_a of 6.7 to 6.9 MeV. Despite some similarities with the Th halo there is no known a-decay sequence corresponding to these energies. Although well documented (and even photographed) by Joly, Gentry has yet to find any X halos in scanning thousands of sections of biotite from Ytterby, Arendal, and Capetown. Above all others, the search for this halo has taxed my eyesight. In this respect the much earlier (and quite obscure) reports (Schintimeister et al 36) of genetically related a-decay of 3 MeV and 4.5 MeV are interesting, but they remain unconfirmed. Therefore any association with the very elusive X halo is only speculation.

Gentry (37) has reported the existence of a halo with rings apparently due to a-energies of about 4.4 to 5.4 MeV. However, the relatively large size of the inclusion of this halo (6 mm in diameter) necessitates a re-examination of this halo with other techniques. Possibly the inner disc is a ghost ring resulting from a-particle attenuation within the inclusion. If so, the halo may be the ^{210}Po variety. I have also reported a halo possibly due to ^{211}Bi a-decay (30). Thus far I found only two of these halos and more specimens are needed to confirm the identity of this type.

Another unusual halo was the so-called *D* halo reported by Henderson (32) to exhibit a diffuse boundary of radius ~16 mm. He tentatively attributed this halo to ^{226}Ra a-decay because the radius approximated the size of the ^{226}Ra ring in the U halo. The absence of other rings, which should have appeared from daughter product a-activity, was explained by assuming ^{222}Rn diffused from the inclusion before it decayed. This, of course, is contrary to the situation observed in the normal ^{238}U halo. Gentry has examined such halos, both in Henderson's original thin sections and in other biotites. They generally possess inclusions several microns in diameter and are without detailed ring structure. While they cannot be explained on the basis of ^{226}Ra a-decay, the presence of fossil fission tracks indicates that U series cc-emitters produced part of the coloration in this halo type.

In addition to U and Th halos Iimori & Yoshimura (18) reported three halo sizes they designated as Z_1, Z_2, and Z_3 halos. These halos were attributed to actinium series a-emitters. Gentry has examined some of the original slides as well as separate Japanese biotite samples in which such halos were reported. Gentry has observed that many halos in this biotite are very dark, so that it is necessary to prepare very thin sections in order that the halo inclusion can be seen. Some of the original sections containing Z halos were simply too thick to permit accurate halo radii measurements. In my opinion the Z_1 and Z_2 halos can be explained as U and/or combination U-Th halos without postulating the actinium a-emitters. The Z_3 halo is actually a ^{210}Po halo. In this context it should be noted that Joly's "emanation" halo was also a ^{210}Po halo.

Giant Halos and Unknown a-Radioactivity

Although Hirschi (21) was apparently the first to report giant halos, Wiman's (17) report of biotite halos with radii of *55* mm and 67 mm was better documented. Even so, on the basis of a rather cursory examination, Hoppe (38) was unable to confirm Wiman's results. Gentry (8), however, later found, after examining more than 1000 thin sections, that giant halos do exist in the rocks described by Wiman.

A more abundant source of giant halos was found in a mica from Madagascar. In this specimen Gentry (8) reported seven different groups of giant halos ranging from 45 mm to 110 pm. An unanswered question is whether these halos originate with high energy a-emitters in the range from 9.5 MeV to 15 MeV. One group in particular with radii between 50 and 58 mm was tentatively attributed to the low-abundance (1:5500), high energy (10.55 MeV) a-particles of ^{212}Po, the last a-emitter in the Th decay chain. Tentatively, this low-abundance group was associated with the 55-mm radius giant halo in the granites that Wiman studied. No other known nuclides occur with sufficient energy and/or abundance to produce the other groups of giant halos.

Therefore, seven other possibilities were considered (8) as explanations for the giant halos. These were: *(a)* Variations in a-particle range due to structural changes in mica, *(b)* Diffusion of a pigmenting agent from the inclusion into the matrix, *(c)* Diffusion of radioactivity from the inclusion to the matrix, *(d)* Channeling, *(e)* b-radiation instead of a-emission, *(J)* Long-range a-particles from spontaneous fission, and (g) a-particles or protons from *(n, a)* or (a, p) reactions.

Gentry (8) has shown that none of the above alternatives are very probable, implying that the giant halos may well represent unknown a-radioactivity. In this respect the giant halos in cordierite reported (and photographed) by Krishnan & Mahadevan (39) are very significant. The "Th" giant halos they report may be due to the low abundance as from ^{212}Po. In contrast the "U" giant halos correspond to an E_a of about 9.5 MeV and cannot be explained on the basis of a low212superheav5 elem

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very light Th halos. Figure 2*h* exhibits a giant halo of about 90-mm radius. If produced by high energy as, the halos in Figure 2*g*, *h* correspond to an E_a of 11.6 and 14 MeV respectively.

Mass Analysis of Halo Inclusions

The preceding sections have presented data on unusual size halos that may well have important implications for nuclear science and cosmology. If, for example Po halos did originate with isomers that were long-lived *b*-decaying precursors of Po, then some of these isomers may represent extinct natural radioactivity (Kohman 41) and hence will be of cosmological significance. Possibly the isomer hypothesis for the giant halos may bear similar implications. Now the above inferences have been deduced solely on the basis of halo ring structure. Clearly the variant halo inclusions themselves contain another important source of information about the radionuclides which generated such halos. However, the small inclusion size effectively prevents use of ordinary mass spectrometric techniques.

To circumvent these problems Gentry has utilized the recently developed ion microprobe mass spectrometer (Anderson & Hinthorne 42) for in situ mass analysis of even the smallest halo inclusions (8, 28, 40). Many U and Th inclusion were analyzed to obtain isotopic data against which the results from the variant halos may be compared. The most important specific application thus far has been the analysis of Po halo inclusions.

Because the Po halos shown in Figure 1*p*, *q*, *r*, *s*, *t* all initiate with Po isotopes that terminate with ^{206}Pb, these inclusions should reflect an excess of this Pb isotope; further because ring structure and fission track analysis show only small amounts, if any, of U, mass analysis of such Po halo inclusions should be consistent with these observations as well. A mixed type Po halo such as in Figure 1*u* (or Figure 1*o*) would be expected to exhibit excesses of both ^{206}Pb (from ^{210}Po decay) and ^{208}Pb (from ^{212}Po decay). If ^{211}Bi halos have been properly identified (30), an excess of the decay product ^{207}Pb should exist in these inclusions. Generally then, while any given Po halo inclusion might have initially contained varying amounts of the different Po isotopes (or their *b*-decaying precursors), halo rings would develop only if ~10^8 atoms of a specific nuclide were present. This implies Po isotope ratios may be variable even when examining different Po halos of the same general type. (In principle this is similar to the case where varying amounts of Th are found in halo inclusions around which only U rings appear.)

206 P206decayinge Pbee 2 Po de6

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