Superheavy Elements

Proceedings of the International Symposium
on Superheavy Elements
Lubbock, Texas — March 9-11, 1978

Editor
M. A. K. Lodhi

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SYMPOSIUM PROGRAM

1. All sessions will be held in the Senate Room University Center, Texas Tech University.
2. Each contributed paper will be given ten miuntes for oral presentation and the time for the invited talk is variable, it is therefore indicated within the parenthesis against the name of the invited speaker.

WEDNESDAY, MARCH 8, 1978

5:00 p.m.-9:00 p.m. Registration, Lobby, Lubbock Inn

7:30 p.m.-9:30 p.m. Informal get together-Meeting Room, Lubbock Inn

THURSDAY, MARCH 9, 1978

8:00 a.m.-1:00 p.m. Registration, Lobby University Center by Senate Room.

8:30 a.m.-9:30 a.m. SESSION I: Opening of the Symposium
Chairman: W.O. Milligan, Welch Foundation

Opening Remarks: M.A.K. Lodhi, Symposium Chairman

Welcoming Address: On behalf of the Texas Tech Univeristy

Inaugural Lecture: "Overview: History and Perspective of the Search for Superheavy Elements."
O. Lewin Keller

Coffee Break

9:45 a.m.-12:30 p.m. SESSION II: Searches for SHE at Accelerators
Chairman: A. Ghiorso, Berkeley

1. "Attempt to Produce SHE in Reactions between Very Heavy Nuclei"
G. Herrmann (30 min)

2. "Recent Searches for Superheavy Elements"
E.K. Hulet (20 min)

3. "Experimental Prospects for the Synthesis and Detection of Superheavy Elements"
J.M. Nitschke (20 min)

4. "New Experimental Insights into the Production of Superheavy Elements Using Heavy Ion Reactions"
R.J. Otto, D.J. Morrissey, D. Lee, A. Ghiorso, G.T. Seaborg, W.D. Loveland, and M. de Saint-Simon

5. "Production of Actinides and Evidence for the Possible Production of SHE via Secondary Reactions in the Bombardments of Tungstens with 24 GeV Protons"
A. Marinov

6. "Neutron Multiplicity Measurements for Spontaneous Fission of Cf and Fm Isotopes and Relevance to Neutron Emission for SHE"
D.C. Hoffman (15 min)

7. "Long Range Fission Fragments from Radiogenic Lead"
J.A. Maly and D.R. Walz

2:00 p.m.-5:00 p.m. SESSION III: Quest for SHE in Nature
Chairman: E.M. McMillan, Berkeley

1. "Search for Superheavy Elements in Nature (Meteorites) and in Heavy-Ion Reactions"
R. Brandt (30 min)

2. "Observation of Anomalous Long-Range Alpha Particles and Their Possible Connection to Superheavy Matter"
S.N. Anderson, J.J. Lord, R.J. Wilkes, J.Albers, L. Barrett, P. Kotzer, R. Lindsay, K. Stehling

→ 3. "Are Any Unusual Radio Halos Evidence for SHE?"
R.V. Gentry (30 min)

Coffee Break

4. "Existence of New Long-Range Alpha Particles in Nature"
A. Chevallier, J. Chevallier, A. Pape, M. Debeauvais, B. Leroux

5. "Analysis of a Giant Halo Monazite Inclusion on the Harwell Proton Microprobe"
J.A. Cookson, N.R. Fletcher, K.W. Kemper, L.R. Medsker, T.A. Cahill

6. "Superheavy Elements in the Early Moon"
S.K. Runcorn

7. "Super-giant Halos in Madagascar Biotite"
C.A. Grady, R.M. Walker

8. "Comments on the Recent Reports of SH Elements in Meteorites and Hot Springs Extracts"
G. Flynn, P. Fraundorf, J. Shirck, R.M. Walker

9. "Experimental Constraints on the Existence of Superheavy Elements in Allende Acid Residues"
G. Flynn, P. Fraundorf, J. Shirck, R.M. Walker, C. Stephan

6:30 p.m. Symposium Banquet at the Lubbock Club, Downtown.
After-dinner speaker, Dr. Richard C. Atkinson, Director, National Science Foundation, USA

Title: "U.S. Science--A Current Perspective"

FRIDAY, MARCH 10, 1978

9:00 a.m.-12:30 p.m. SESSION IV: Theoretical Predictions for SHE
Chairman: M. Goldhaber, Brookhaven

ARE ANY UNUSUAL RADIOHALOS EVIDENCE FOR SHE?

Robert V. Gentry†
Oak Ridge National Laboratory*, Oak Ridge, Tennessee 37830

ABSTRACT

Of the ∿10^5 radiohalos examined thus far (out of an estimated >10^{18} in the earth's crustal rocks), the great majority can be associated in some way with the α-decay of ^{238}U and/or ^{232}Th. In addition there are a significant number of medium size halos which can be attributed to the α-decay of several isotopes of Po. Finally, there is a residue of a few halos of large, medium, and small dimensions that have thus far proved difficult to associate with any known type of α-radioactivity. While evidence for various theories concerning the origin of these halos is discussed in the following article, more evidence is needed to determine whether SHE were (or are) involved in their genesis.

INTRODUCTION

Ordinary radiohalos are herein defined as those which initiate with ^{238}U and/or ^{232}Th α-decay (1), irrespective of whether the actual U or Th halo closely matches the respective idealized α-decay patterns. In a relatively few instances the match is very good. Compare for example the idealized U halo ring pattern in Fig. 1a with the well developed U halos in biotite (Fig. 1f) and fluorite (Fig. 1h,h'); these halos have ring sizes that agree very well (2) with ^{4}He ion accelerator induced coloration bands in these minerals (see Table 1). In these cases a halo ring can be assigned to a definite α-emitter.

In other cases, however, such as the halos in fluorite (2,3) shown in Fig. 1g,i-m, much work was required before these halos could be reliably associated with U α-decay (2). As explained elsewhere (2), reversal effects accompanying extreme radiation damage caused the appearance of rings that could not be associated with definite α-emitters of the U decay chain. We conclude then that many halos may exhibit a ring structure different from the idealized U and/or Th α-decay patterns because of reversal effects. Experience in examining many halos (∿10^5) has also shown that in spite of the fact that most of the >10^{18} halos in earth's crustal rocks have blurred or obliterated ring structure due to the large size of the inclusions, nevertheless the outer dimensions of most of these halos allow them to be readily classified as U and/or Th types.

Of the four types of unusual halos that appear distinct from those formed by U and/or Th α-decay, only the polonium halos (see Fig. 1b-d,n-r,r') can presently be identified with known α-radioactivity (2-4). Because it is impossible to ascertain from ring structure analysis alone whether any of the other three unusual types, viz., the X, dwarf, or giant halos (5), were formed by superheavy element (SHE) decay, other techniques have been used to explore this possibility. In particular, the proton induced x-ray fluorescence (PIXE) experiments (6), which initially suggested an association of giant halos (GH) with SHE of $Z = 126$, were not confirmed by subsequent synchrotron radiation x-ray fluorescence (SRXRF) techniques (7,8). (See Fig. 2 for a comparison of the original PIXE data on GH inclusion 19D with the results of the SRXRF experiments, and References 7 through 10 for further explanation of the data.)

GIANT HALOS IN MADAGASCAN MICA

Review

Previously eight conventional hypotheses were considered as possible explanations (5) of GH. These were: (a) enhanced α-particle range resulting from structural or physical changes in

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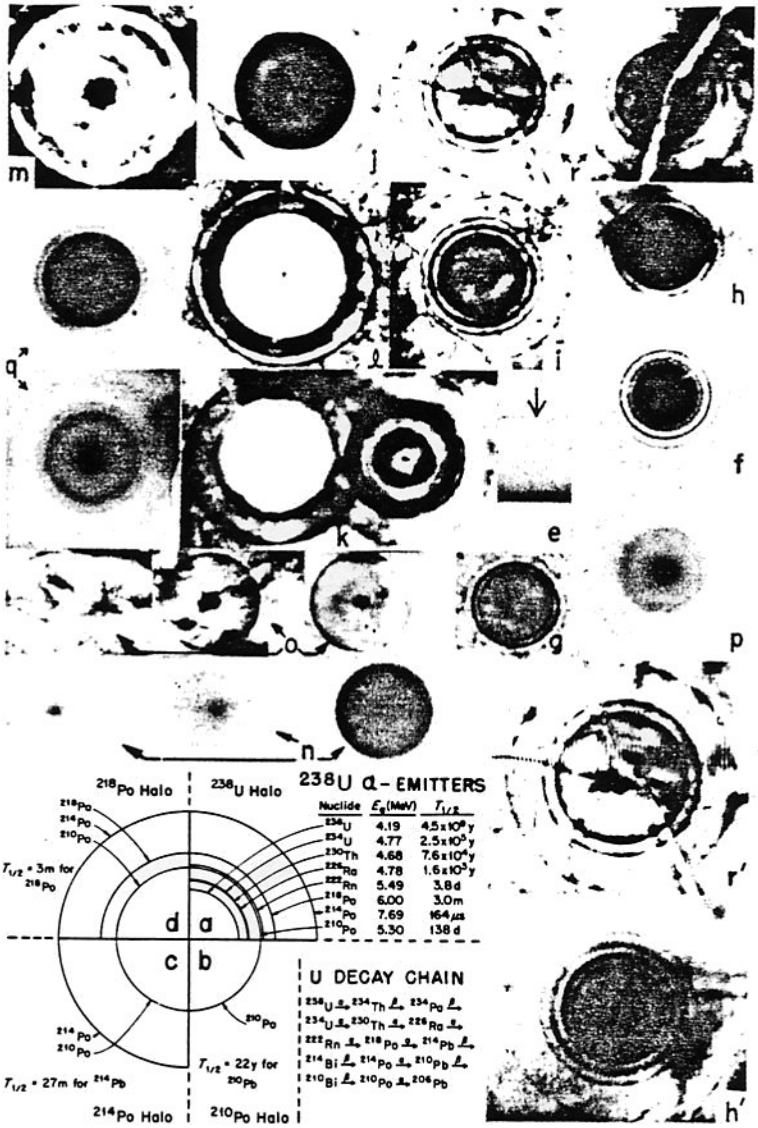
*This research was supported by the Division of Nuclear Sciences, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation.



^{238}U α- EMITTERS

Nuclide	E_α(MeV)	$T_{1/2}$
^{238}U	4.19	4.5×10^{9} y
^{234}U	4.77	2.5×10^{5} y
^{230}Th	4.68	7.6×10^{4} y
^{226}Ra	4.78	1.6×10^{3} y
^{222}Rn	5.49	3.8 d
^{218}Po	6.00	3.0 m
^{214}Po	7.69	164 μs
^{210}Po	5.30	138 d

Fig. 1. The scale for all photomicrographs is 1 cm ≈ 30.0 μm, except for (h') and (r'), which are enlargements of (h) and (r). (a) Schematic drawing of ^{238}U halo with radii proportional to ranges of α-particles in air. (b) Schematic of ^{210}Po halo. (c) Schematic of ^{214}Po halo. (d) Schematic of ^{218}Po halo. (e) Coloration band formed in mica by 7.7-MeV ^{4}He ions. Arrow shows direction of beam penetration. (f) A ^{238}U halo in biotite formed by sequential α-decay of the ^{238}U decay series. (g) Embryonic ^{238}U halo in fluorite with only two rings developed. (h) Normally developed ^{238}U halo in fluorite with nearly all rings visible. (h') Same halo as in (h) but at higher magnification. (i) Well-developed ^{238}U halo in fluorite with slightly blurred rings. (j) Overexposed ^{238}U halo in fluorite, showing inner ring diminution. (k) Two overexposed ^{238}U halos in fluorite showing inner ring diminution in one halo and obliteration of inner rings in the other. (l) More overexposed ^{238}U halo in fluorite, showing outer ring reversal effects. (m) Second-stage reversal in a ^{238}U halo in fluorite. The ring sizes are unrelated to ^{238}U α-particle ranges. (n) Three ^{210}Po halos of light, medium, and very dark coloration in biotite. Note the differences in radius. (o) Three ^{210}Po halos of varying degrees of coloration in fluorite. (p) A ^{214}Po halo in biotite. (q) Two ^{218}Po halos in biotite. (r) Two ^{218}Po halos in fluorite. (r') Same halo as in (r) but at higher magnification.

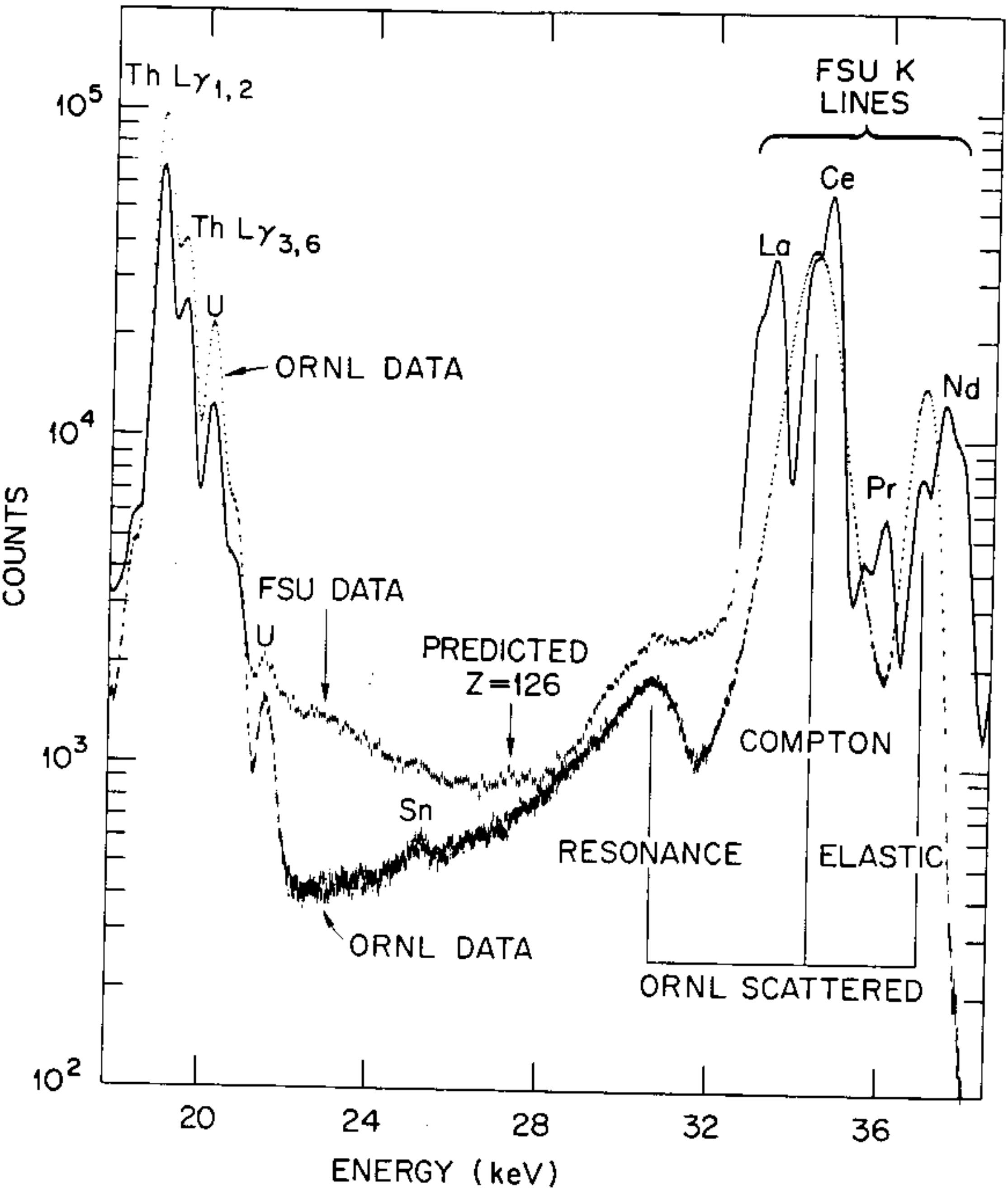


Fig. 2. Comparison of proton induced (FSU) and synchrotron radiation (ORNL) x-ray fluorescence data on giant halo inclusion 19-D

Table 1. Comparison of sizes of induced bands (columns 1 to 5) with halo radii (columns 8 to 21). Column 6 gives the ^{4}He ion energies at which the induced bands were formed, or the α-particle energies corresponding to the nuclides in column 7. Thus, the nuclide or α-particle energy that produced any halo ring in columns 8 to 21 can be found from column 6 or 7. The letters K-L, H, S, M, and G represent halo measurements by Kerr-Lawson (*15*), Henderson (*1, 6, 7*), Schilling (*9*), Mahadevan (*10*), and Gentry. Subscripts L, M, and D indicate light, medium (dose 10 to 20 times coloration threshold), and dark (dose about 50 times coloration threshold) induced bands; L → D and L → M indicate light to dark and light to medium; these were visually determined. Gentry's measurements were made with a filar micrometer readable to 0.07 μm. The estimated overall uncertainty was ± 0.3 μm. Other abbreviations: N.M., not measured; N.R., not resolved; N.P., not present.

Coloration band size (μm)							U halo radius (μm)						Po halo radius (μm) in							
Biotite			Fluorite	Cordierite	E (Mev)	Nuclide	Biotite			Fluorite		Cordierite	Biotite				§		Fluorite	
													^{210}Po		^{214}Po		^{218}Po		^{210}Po	^{214}Po
1. G_L	2. G_M	3. G_D	4. G	5. G	6.	7.	8. K-L	9. H	10. G	11. S	12. G	13. M	14. H	15. $G_{L\to D}$	16. H	17. $G_{L\to M}$	18. H	19. G_M	20. G	21. G
13.4	13.8	14.2	14.1	16.2	←4.2	^{238}U→	12.3	12.7	12.2→13.0	14.0	14.2	16	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.
N.M.	16.7	N.M.	17.3	19.2	←4.77	^{226}Ra→	15.4	15.3	14.8→15.8	16.9	17.1	19	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.
N.M.	N.M.	N.M.	N.M.	N.M.	←4.66	^{230}Th→	N.R.	N.R.	N.R.	15.8	N.R.	N.R.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.
N.M.	16.7	N.M.	17.3	19.2	←4.78	^{234}U→	15.4	15.3	14.8→15.8	16.9	17.1	19	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.
N.M.	19.3	20.0	19.6	22.5	←5.3	^{210}Po→	N.R.	N.R.	N.R.	19.3	19.5	N.R.	19.8	18.3→19.9	20.0	18.1→19.1	19.9	19.3	19.8	19.8
N.M.	20.5	21.1	N.M.	N.M.	←5.49	^{222}Rn→	18.6	19.2	18.1→19.0	20.5	20.5	23.5	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.	N.P.
N.M.	23.0	23.9	23.6	26.7	←6.0	^{218}Po→	22.0	23.0	21.5→22.7	23.5	23.5	26.5	N.P.	N.P.	N.P.	N.P.	24.0	23.3	N.P.	23.7
33.1	33.9	34.4	34.6	38.7	←7.69	^{214}Po→	33.0	34.1	30.8→33.0	34.5	34.7	38.5	N.P.	N.P.	34.5	32.5→33.8	34.0	34.0	N.P.	34.9

the mica adjacent to the inclusion, (b) diffusion of some chemical from the inclusion into the matrix, (c) diffusion of radioactivity from the inclusion into the matrix, (d) channeling, (e) β-radiation instead of α-emission, (f) long-range αs from spontaneous fission of ^{238}U, (g) α-particles or protons from (n,α) or (α,p) reactions on the inclusion or matrix, and (h) low abundance, high energy αs from ^{212}Po and ^{214}Po in the ^{232}Th and ^{238}U decay chains respectively. Two other more exotic explanations considered were high energy αs from SHE or from unknown isomers of known elements.

Since this earlier study was published three other reports (11-13) have appeared suggesting that GH may have formed from mechanical separation of the mica (11), or from high energy αs from ^{244}Pu (12) fission, or from α-proton knock-on events (13), which requires postulating water in the inclusion. In addition Chevallier et al. (14) report they have found evidence of high energy αs in Madagascan monazite from the Col du Manangotry region and suggest they may have some relation to the GH which were found in the same area. An evaluation of this hypothesis must necessarily await the identification of the source of the high energy αs by these investigators.

Because additional data might turn any one of the either previously or newly suggested explanations into a viable one, or otherwise suggest a completely new possibility, a detailed re-examination of GH was carried out using Ion Microprobe Mass Spectrometer (IMMA) techniques. This research with the ion microprobe has been a collaborative effort with W. H. Christie and D. H. Smith of the Analytical Chemistry Division at ORNL and with S. S. Cristy and J. F. McLaughlin of the Laboratory Development Division, Y-12 Plant, Oak Ridge, Tennessee.

Giant and U-Th Halos with Opaque Inclusions

The most definitive and as yet unexplained result of this re-analysis was obtained in searching for differences in elemental and isotopic abundances between the U-Th and GH halo inclusions in the Madagascan mica. That is, if GH had formed from unknown high energy isomers α-decaying to Pb (5), or from high energy αs from ^{244}Pu spontaneous fission (12), then in the first case GH inclusions might reveal a corresponding Pb isotopic enrichment while in the second case a Th enrichment might be evident when compared to the U-Th halo inclusions. While the idea looks good in theory, in practice the experiments suffer from the fact that we are trying to detect small differences in large Th and Pb secondary ion signals obtained from the U-Th and giant halo inclusions. Quite importantly, however, these experiments did lead to the discovery that certain optically opaque inclusions, which are mineralogical oddities possessing no external crystalline form and which occur somewhat rarely as the radiocenters of both U-Th and giant halos, consistently exhibit marked deficiencies in their ^{208}Pb content compared to the much more abundant monazite halo inclusions. Before elaborating further on this discovery, we first discuss the experimental results which pertain directly to various explanations advanced for the origin of the GH.

Evaluation of Possible Explanations for Giant Halos

In particular the PIXE (6,9,10), SRXRF (7,8) and IMMA results have shown approximately the same relative abundances of U, Th, and Ce for many of the GH and U-Th monazite inclusions. Since Ce is one of the major elemental constituents (∿15-20%) of monazite, this means that, at least in the halos investigated, the overall abundance of U and Th does not vary significantly between U-Th and GH monazite inclusions. (IMMA results, however, have shown spatial variation of for example Th within the inclusions themselves.) Thus all theories of GH origin which require an abnormally high concentration of α-radioactivity in the GH inclusions are now rendered untenable unless there exists another unknown source of α-activity.

For example, in addition to the fact that many of the GH in the Madagascan mica referred to herein are both significantly smaller and more sharply delineated than the rather diffuse boundary β-radiation produced halos reported in quartz (15), we can now virtually eliminate the β-ray hypothesis because of the comparable abundance of the most significant β-activity (U and Th decay chains) in the U-Th and GH monazite inclusions. Likewise, contrary to an earlier inference (5), the GH with 55-60 μm radii can no longer be associated with an excessive number of low abundance 10.55 MeV αs from ^{212}Po in the ^{232}Th series. In my opinion, the only way in which high energy αs from ^{212}Po could possibly have remained an option (16) for GH formation was for ^{212}Po to have been incorporated into the GH inclusions independent of ^{232}Th. However, the fact that IMMA mass scans of GH inclusions showed no enrichment of ^{208}Pb, the decay product of ^{212}Po, means that there is no evidence to support this hypothesis.

The idea that GH could have formed from low abundance high energy αs from spontaneous fission of extraordinary amounts of ^{238}U in a GH inclusion has very little credibility, first because the fission decay mode is only $\sim 5\times 10^{-7}$ that of ^{238}U α-decay, second because high energy αs are emitted in only about 1:400 fission events, and third because, even if enough αs were available, the Gaussian distribution (which peaks at ~16 MeV) of these αs is so broad (~8 MeV at FWHM) that it is difficult to conceive how such conditions could produce a well defined halo periphery. In my opinion the added data showing relative equivalence of U between U-Th and GH inclusions erases any lingering doubts about the credibility of the ^{238}U fission-α hypothesis.

In this context we should note some of the problems involved in accepting the idea that GH might have formed from high energy αs associated with ^{244}Pu fission (12). Even though the fission vs α-decay mode is more favorable (12) for ^{244}Pu (λ_f/λ_α=1:800) than for ^{238}U ($1:2\times 10^6$), still the objections cited above about the low frequency of high energy α-emission in a fission event and the extended diffuse halo periphery expected from such αs are just as damaging to the credibility of the ^{244}Pu fission hypothesis as they are to the idea involving ^{238}U fission.

The possibility that αs from (n,α) reactions could have caused these GH was earlier (5) rather convincingly tested by simply exposing mica sandwiches with halo inclusions to a high neutron fluence. No coloration was observed with a fluence of $>10^{17}$ neutrons/cm^2, which is orders of magnitude higher than the fluence received by the host mineral since crystallization. In contrast to this experimental test about the (n,α) hypothesis, the question of GH production from secondary (α,p) nuclear reactions could only previously be shown to be of minimal probability from the theoretical standpoint. However, the new data on U and Th equivalence in GH and U-Th monazite inclusions assures us that no enhanced α-activity from either of these sources could have caused the GH via this reaction.

By this same data we are likewise assured that channeling is a very improbable explanation for GH. Formation of GH from channeling of necessity has a very small credibility quotient because it requires GH to form from only that extremely small fraction of αs which emanate from the inclusion nearly parallel to the basal cleavage plane of the mica. Thus to obtain the necessary α-fluence to produce coloration from channeled αs required postulating an inclusion with greatly enriched α-activity. In my opinion the U and Th equivalence data has reduced the credibility quotient of this hypothesis to the point where it differs very little from zero.

The question of whether any GH were produced by diffusion of ^{220}Rn($t_{1/2}$=56s) and/or ^{222}Rn($t_{1/2}$=3.8d) out of the GH inclusion was previously tested first by examing the surface of the mica adjacent to the inclusion with autoradiographic techniques. Because of the 30-40 μm range of the α-particles from the Rn daughters, these experiments with Kodak NTA emulsion effectively sampled the entire thickness of the GH and showed that α-radioactivity was presently confined to the site of the inclusion. These results admittedly did not rule out the possibility that the postulated diffused α-radioactivity may have produced an enlarged halo and long since decayed away. The crucial test for the hypothesis is whether any Pb, the decay product of Rn, exists in significant enough quantities in the regions immediately adjacent to the GH inclusions. Previously the electron microprobe was used to search for this Pb in these areas (5), but none was found. Admittedly, however, this technique is relatively insensitive so the question was not fully resolved.

Further experiments were therefore undertaken using the IMMA, an instrument which has been shown capable of detecting as few as $\sim 10^9$ atoms of Pb in a mica matrix. In planning these experiments it was decided that a complete mass scan should be taken of three different regions, viz, (i) within ~10-15 μm of the GH inclusion, (ii) ~10-15 μm inside the outer periphery of the GH, and (iii) ~10-15 μm outside the GH. Figure 3a-c shows a set of the three such mass scans taken within and immediately adjacent to a GH. These mass spectra and others like them taken near other GH make it clear first of all that there are no elemental variations in the mica itself in these three regions and secondly that no ^{206}Pb or ^{208}Pb signal is observed. In my opinion this is very strong evidence that these GH did not originate from Rn diffusion. Indeed, since the size of the inclusion does not determine the size of the halo, on qualitative grounds alone it is quite difficult to understand why diffusion would produce GH around some inclusions and not others. Further the sharply defined peripheries of many GH are clearly incompatible with the diffusion hypothesis. In reality this result is not surprising. As stated above the half-lives for the relevant Rn isotopes are so short that

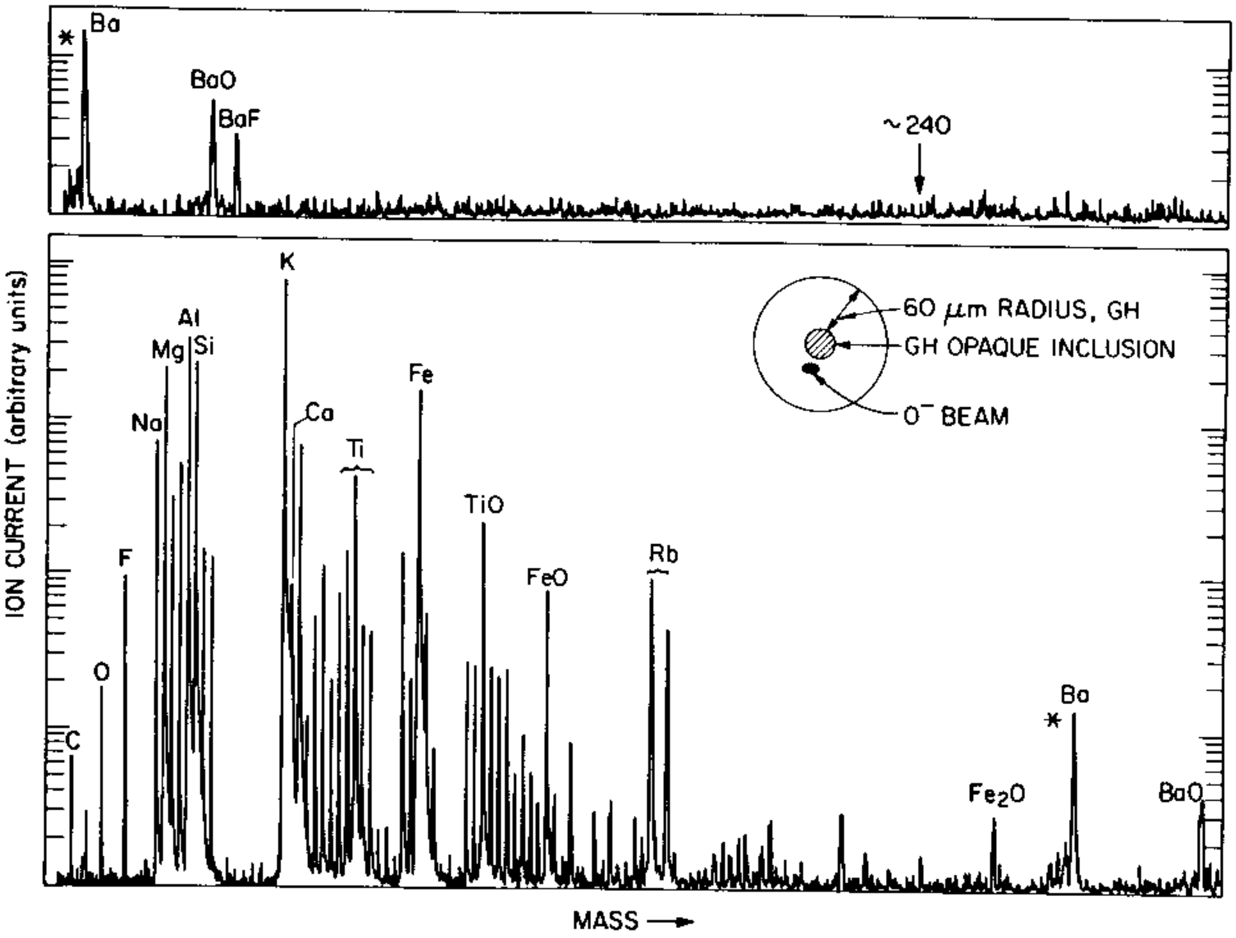


Fig. 3a. Mass scan of Madagascan mica ~10-15 μm from GH inclusion

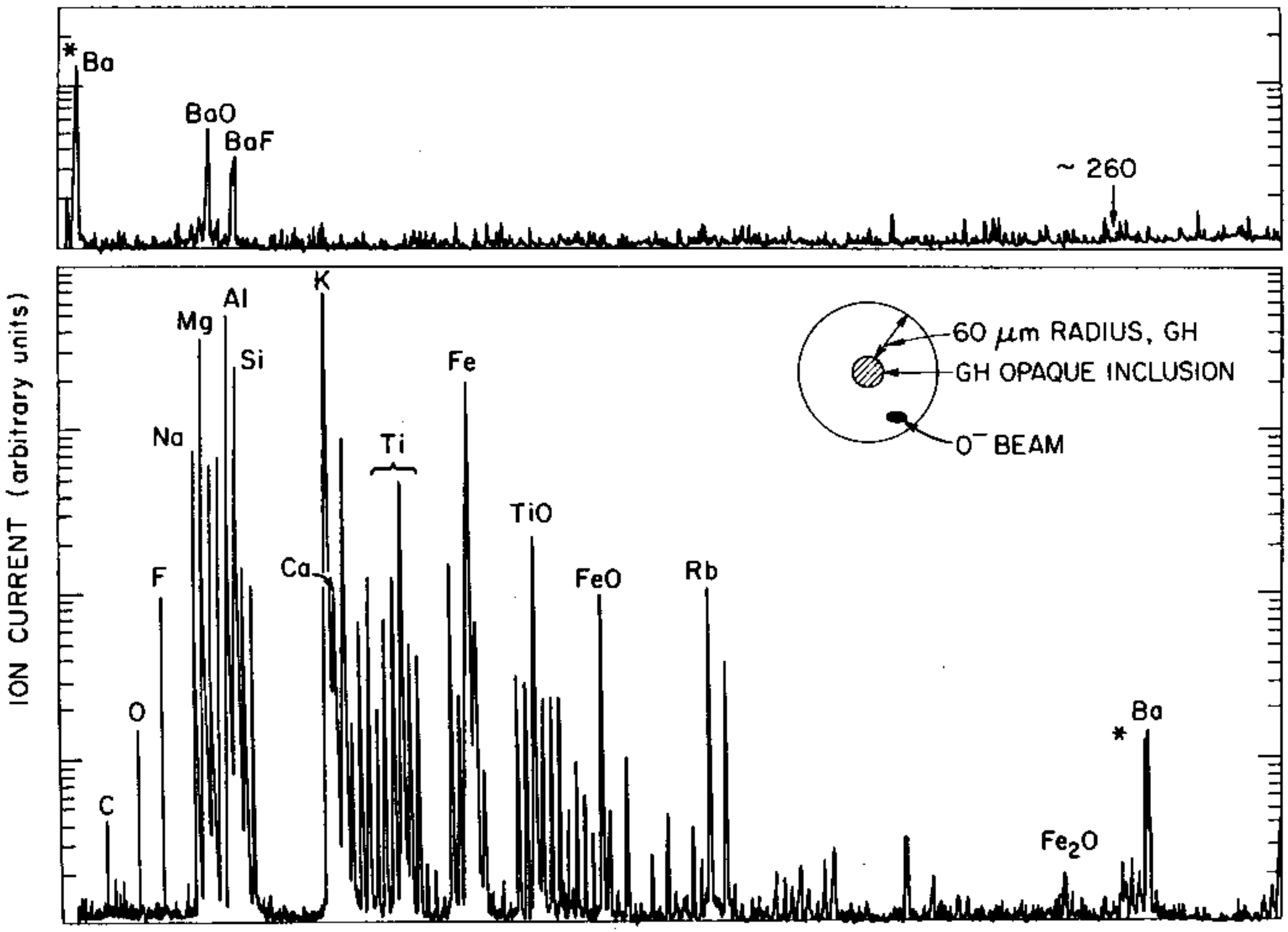


Fig. 3b. Mass scan of Madagascan mica with beam ~10 μm inside GH boundary

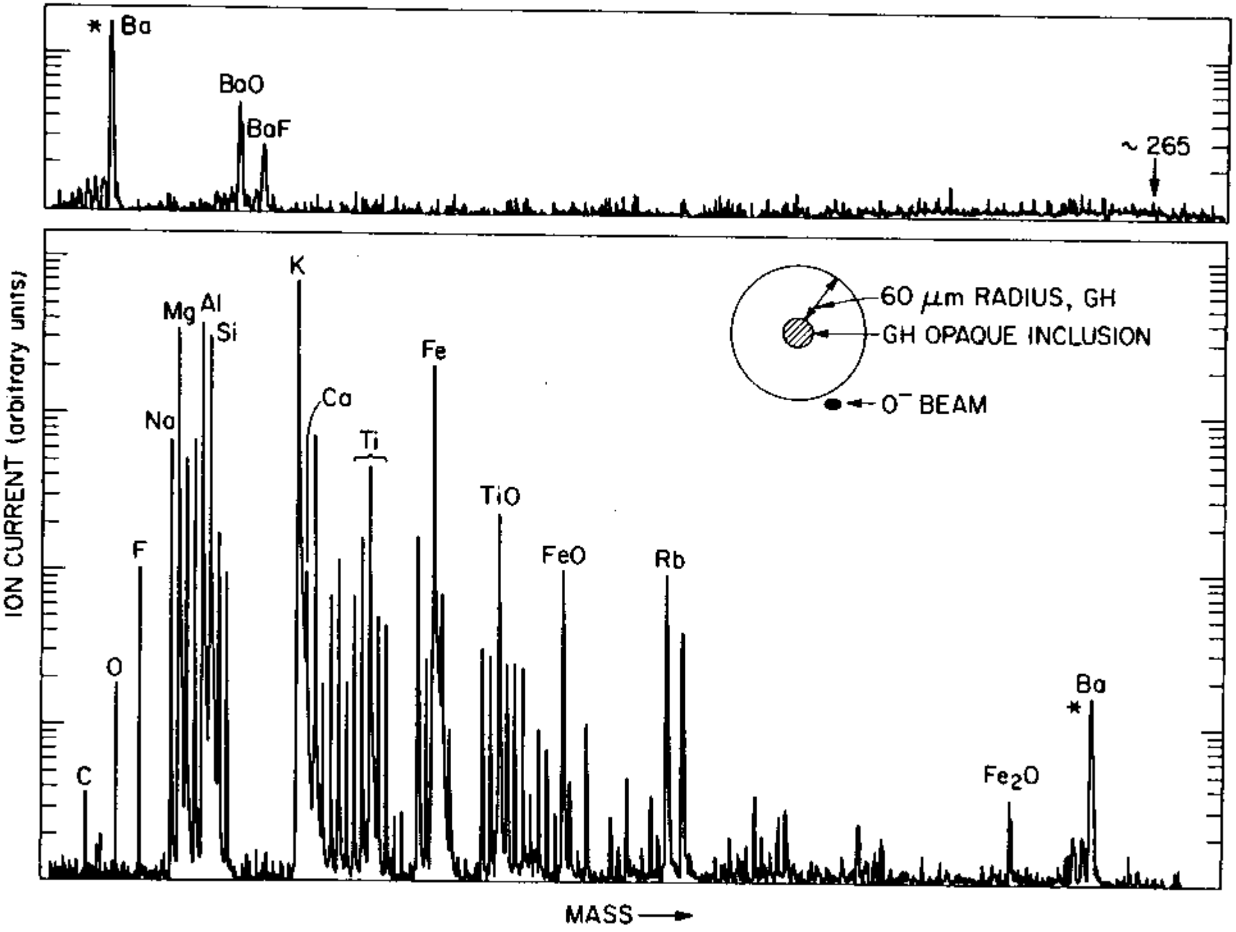


Fig. 3c. Mass scan of Madagascan mica with beam 20 m outside GH boundary

only relatively insignificant quantities would be expected to diffuse out of the inclusion.

As a by-product of these IMMA experiments we were also able to test whether U rather than Rn somehow infiltrated close enough to the inclusion to produce a GH. Notice in Fig. 3 that no trace of U is seen on any of the mass scans. This result correlates well with the HF acid etches which have thus far shown a high density of U fossil fission tracks emanating only from the GH inclusion cavities while only a background density of fossil fission tracks is observed in the halo region itself. The ion microprobe is, by the way, even more sensitive to U than Pb, so that these results can certainly be reasonably interpreted as evidence that only background concentrations (∿ppm) of U exist around the GH inclusions investigated thus far.

In this context we should definitely comment on the idea (11) that Rn, after being liberated from the surface of an inclusion by recoil action, might then diffuse into small crevices in the mica filled with water, or alternatively that normal energy αs emitted from the inclusion might themselves gain enhanced penetration by passing through water filled cracks adjacent to the inclusion. First it is incorrect to conclude that (as was done in Ref. 11) from a single GH photograph (1), which shows a crack around the inclusion, that this necessarily implies cracks are common to all GH and have so existed for extended periods of time. Actually cracks are easily produced if care is not taken when the mica specimen is cleaved for microscopic examination. Cracks appear more frequently around both U-Th and GH inclusions whenever the split reveals an inclusion on the surface and especially when the section is very thin. Cracks are often not seen at all around inclusions in much thicker (∿100 μm) sections that completely enclose the inclusion. Figs. 4-10 show halos with and without cracks.

Nevertheless we have looked for evidence of mica layer separation adjacent to GH inclusions by microscopically examining under high magnification the cavities left in the mica after certain GH inclusions have been carefully extracted. Neither by using this procedure nor by utilizing the classic Newton ring technique (a phenomenon sensitive enough to reveal the extremely small separation (<1 μm) induced by heating effects from a microscope light source) were we able to find definitive evidence of separation of mica layers within the inclusion cavity. This does not invalidate the hypothesis, however, because it was suggested (11) that minute separations might later have annealed.

Now there are some GH with irregular boundaries that do not match the contour of the inclusion and it might seem that here the separation idea has merit. However, the fact that regular size halos also exist with irregular contours suggests that we might be seeing evidence of the same kind of spatial inhomogeneities of U and Th within the inclusion as has been observed with the ion microprobe. Briefly then, the irregular contours of some normal U-Th halos do admit the possibility that the irregular profiles of certain GH could be due to spatial inhomogenieties of some unknown α-emitter. That is, GH with irregular contours do not necessarily have to be explained by the separation hypothesis and in fact there is no firm evidence available that requires this hypothesis as an explanation of GH.

Further, in my opinion there are some GH data which are very difficult to reconcile with this mica separation hypothesis, viz, the sharply defined peripheries of many GH and the apparent discrete distribution of GH. That is, my understanding of the hypothesis (11) is that small cracks or water filled cavities (no trace of which has been found) would be expected to be of variable dimensions. These conditions would seem to imply diffuse halo radii without a discrete size distribution, which is contrary to observations on many GH.

The earlier report (5) that GH exhibited 3-dimensional effects was based on limited observations, one reason being that GH inclusions, which at that time were considered potential sources of SHE, were being erratically lost as the mica containing the GH was split in searching for a 3-D effect. This data is not used in these analyses simply because the results are, in my opinion, not conclusive enough to warrant deductions about specific hypotheses. The main problem is that, in comparison to other micas which generally exhibit *high* differential dark contrast between the halo and the surrounding mica, in this Madagascan mica the halos exhibit *very low* differential *light* contrast with the surrounding mica (halo lighter than mica). That is since the halo inclusions are often >40 μm in diameter, the sections examined are often 60 μm to 100 μm thick and here the total contrast between the halo and the surrounding mica is very good. However, to obtain data on the third dimension the thicker sections containing the GH must be serially sectioned (a process which has resulted in the loss of a number of GH inclusions), and the facts are that for this mica the cleaved sections are often quite thin. But thin sections of halos often show such low contrast with the surrounding mica that sometimes even a section in the plane of the inclusion shows no coloration difference between the halo region and the surrounding mica.

Specifically, serial sectioning of the few (<20) remaining thick (∿100 μm) pieces from the original specimens showed evidence for GH oblate spheroids in six cases and no coloration corresponding to GH dimensions in ten other off-center sections. A partial explanation for these mixed results is that serial sections from thicker mica pieces are sometimes so thin (<5 μm) that the differential color contrast between the halo and the surrounding mica is too low to observe any color difference; thin splits from U-Th halos are observed to show the same effect in this mica. However, it is certain that in some cases the off-center sections were thick enough to observe coloration from GH if the GH had been produced by an α-fluence equal to that of the inner U-Th halo. We have of course no data that would require this equivalence. In my opinion if high energy αs did produce some GH, the α-fluence from this source might even have been sufficient to produce enough contrast so that a GH would be visible when observed in a ∿50 μm thick section without necessarily being observed in a ∿5 to 10 μm off-center section.

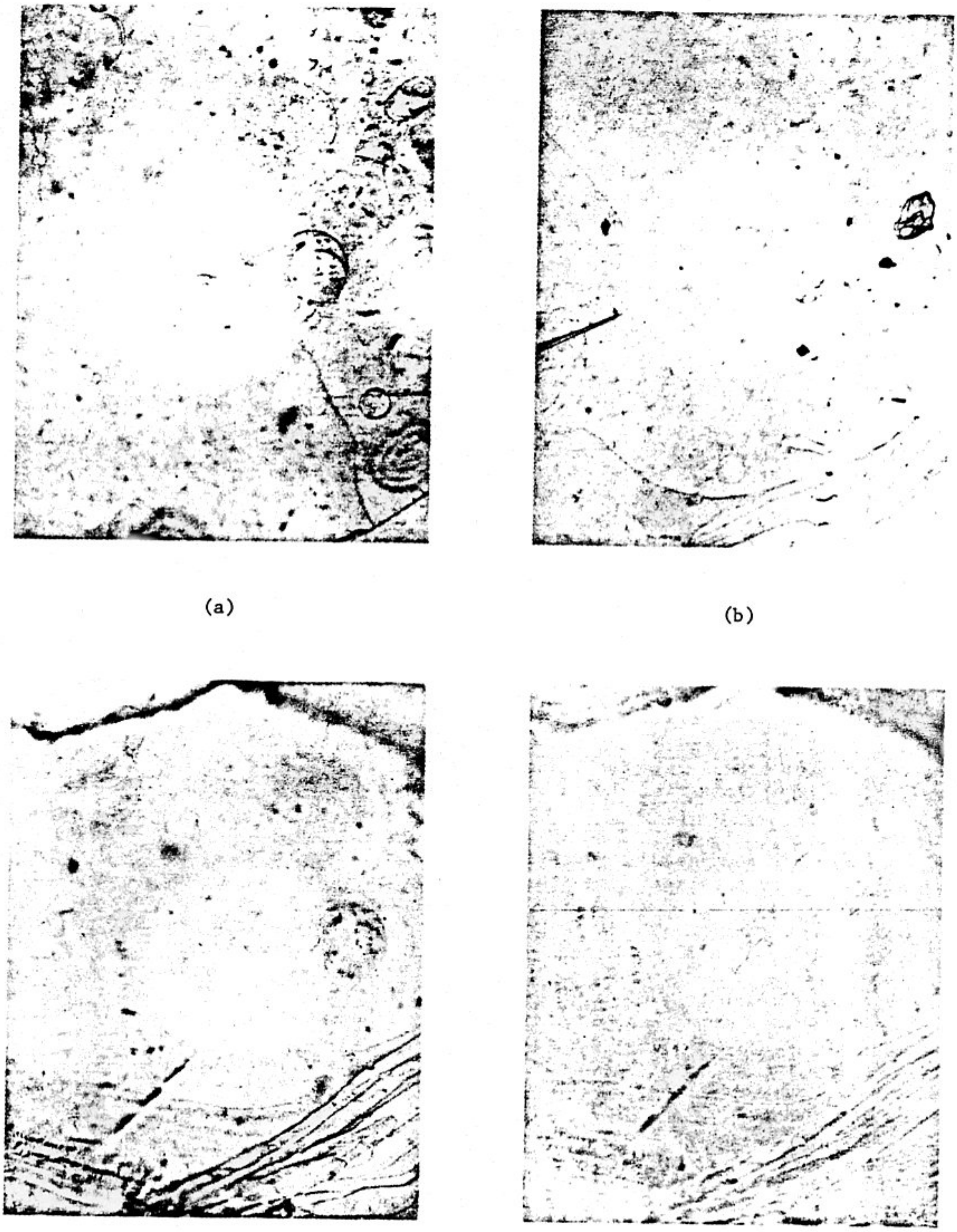
Figure 4(a–d), which shows several ordered serial sections of the halo at increasing distances below a GH inclusion (Fig. 4(a) is a section just a few μm below the inclusion), is illustrative of those GH which exhibit 3-dimensional effects.

An attempt to obtain more information by making transverse sections failed because no halos at all could be observed around the inclusions when this mica was viewed parallel to the basal cleavage plane. Holomicrography was also considered a possibility to gain information without sample destruction, but investigation showed that the resolution of the technique was insufficient to furnish meaningful data.

At this point it is appropriate to present photographic evidence which bears on the question of the size of GH inclusions because this parameter does enter into some ideas advanced for the origin of the GH. Figures 5-10 show a few photographs of U-Th and giant halos in this Madagascan mica. Note that GH and U-Th halos appear in close proximity in three photographs and that some GH do occur around relatively small (∿5 to 10 μm) as well as large diameter inclusions (see Figs. 8 and 9). Cleaving the mica produced cracks around the halos in Figs. 5,7-9.

In my opinion the GH with small inclusions (Fig. 8) raise a serious question about the α-proton knock-on hypothesis because of the drastic reduction in the total number of U and Th α-decays compared to the large size inclusions referred to in Ref. 13. By way of explanation Fig. 2 in Ref. 13 shows that sufficient energy from proton knock-ons was supposedly deposited in the mica to produce coloration out to ∿80 μm from the inclusion. These results were based, however, on the assumption of a 30 μm GH inclusion radius. A reduction in the radius of the GH inclusion from 30 μm to ∿4 μm (see the ∿50 μm radius GH with the ∿4 μm radius inclusion in Fig. 8), reduces the α-fluence from U and Th α-decay by a factor of 4.2×10^2. Also according to Ref. 13, the average area density of H atoms (assuming a homogeneous mixture of monazite and water) encountered by escaping α-particles is proportional to the radius, so that another 7.5 fold reduction is here involved. If my understanding of the calculations in Ref. 13 are correct, then the 420 fold reduction in α-fluence will of itself significantly reduce the distance from inclusion at which the proton fluence would be expected to produce threshold coloration. Instead of the proton fluence inducing coloration in the mica at ∿80 μm from the inclusion as calculated in Ref. 13, there is some question whether the proton fluence would be sufficient to produce coloration even within a few μm from the inclusion. There is little reason to attempt to define just how close to the inclusion proton induced threshold coloration might develop because such small distances are within the U-Th halos, which implies coloration from knock-on protons would not be distinguishable from normal α-induced coloration of the halo. This means that even under the rather liberal assumption of 30% water in a GH inclusion, the U-Th α-fluence from small inclusions is much too low to provide the knock-on proton fluence necessary to produce coloration at distances outside the normal U-Th halos.

Another objection to this hypothesis is that according to Ref. 13 the energy density deposition in the mica due to the proton fluence decreases monotonically with distance from the inclusion. In my opinion the difficulties in understanding how this monotonic decrease could produce halos with sharply defined peripheries, even if enough proton fluence were available, greatly exceeds the problem of explaining how significantly different amounts of water could have been incorporated into U-Th and giant halo inclusions that are as closely spaced as those



(a)

(b)

(c)

(d)

Fig. 4. (a–c) Transmitted light photomicrographs of three off center serial sections of ∿70 μm GH showing dimensions of the GH at increasing distances below the inclusion, (d) same section as (c) but in reflected light.

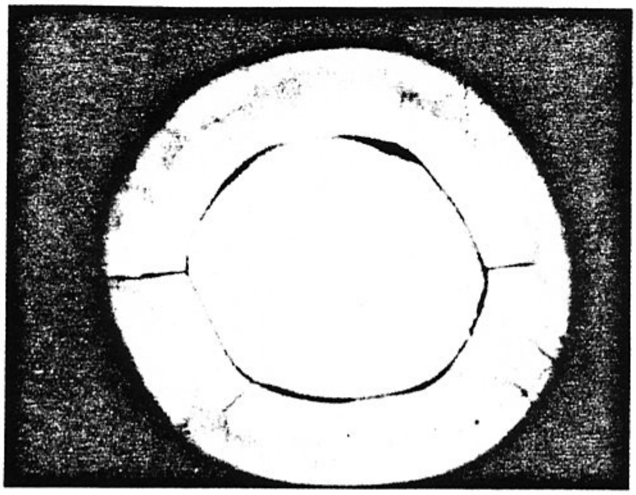


Fig. 5. U-Th halo of ≈40 μm radius with a ≈125 μm diameter inclusion

Fig. 6. U-Th halo of ≈40 μm radius with a ≈7 μm diameter inclusion adjacent to a GH of ≈75 μm radius with a ≈45 μm diameter inclusion

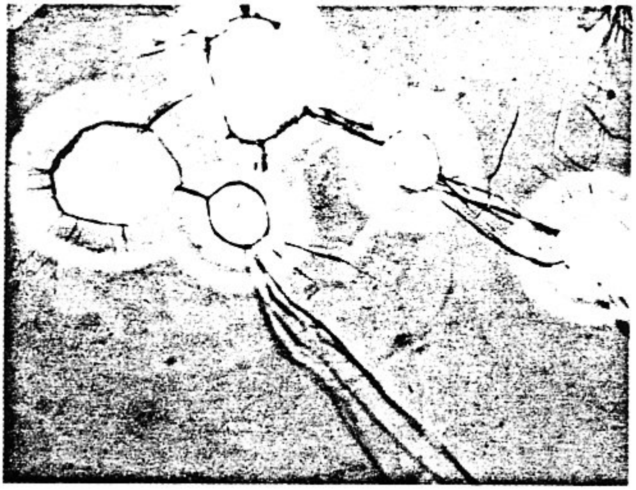


Fig. 7. Group of five U-Th halos with inclusions ranging from about 50 μm to 100 μm in size. Cracks, when they appear around the inclusions as in this photo, are generally produced when the mica is split for microscopic examination.

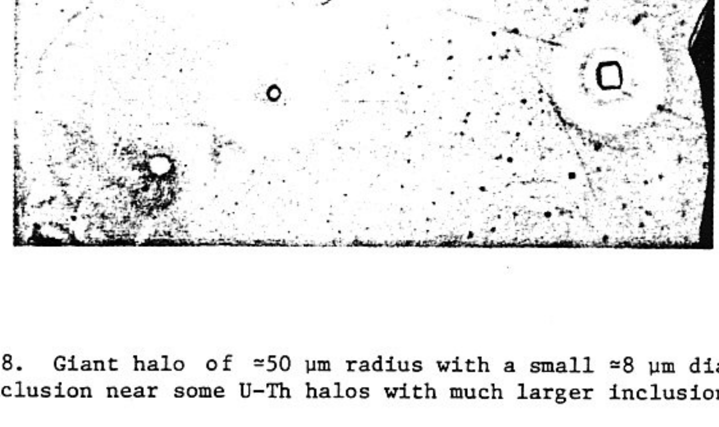


Fig. 8. Giant halo of ≈50 μm radius with a small ≈8 μm diameter inclusion near some U-Th halos with much larger inclusions

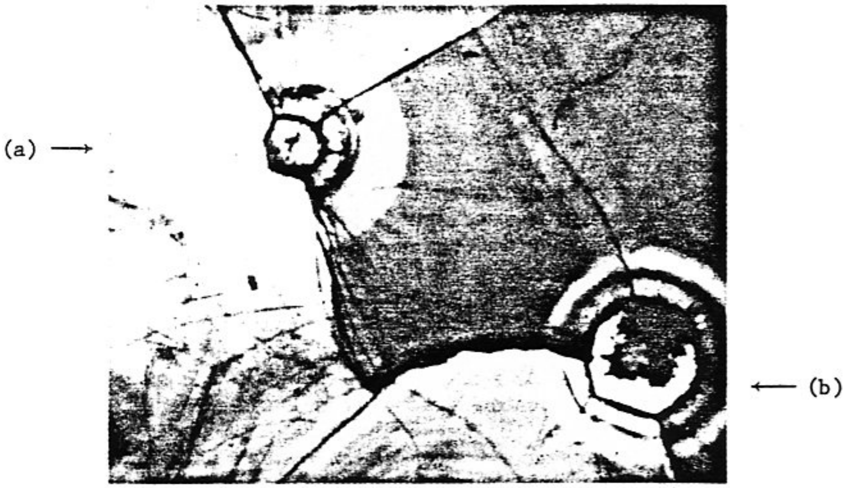


Fig. 9. (a) Giant halo of radius ≈70 µm with a ≈50 µm diameter inclusion, (b) U-Th halo of ≈40 µm radius with a ≈100 µm diameter inclusion. Halos (a) and (b) above are respectively GH 19A and U-Th halo 19B used in the original PIXE experiments (Ref. 6) at FSU.

Fig. 10. Giant halo of ∿70 µm radius with opaque inclusion. See Figs. 12 and 13 for mass spectra of this type of inclusion.

shown in Figs. 6, 8, and 9. This hypothesis also *incorrectly* predicts, because of similar amounts of U and Th in different inclusions, that GH radii would depend on the inclusion size.

Whether any of these GH resulted from normal energy αs passing through a reduced density region (induced by radiation damage effects) requires comment because some GH in Scandanavian micas do exhibit regions adjacent to the inclusion with elemental abundances significantly different than the surrounding mica (see later discussion). However, contrary to the GH in the Scandanavian micas, we have found no evidence of any elemental abundance changes in the regions around the GH in this Madagascan mica (see Fig. 3a-c). We conclude that these GH were not produced via any mechanism associated solely with structural or elemental abundance differences in the region adjacent to the inclusion.

At this point we note that rather infrequently halos of normal size are found with inclusions that contain very small amounts of U or Th. While the coloration of these relatively rare halos (<1:200 U-Th halos) resembles the coloration of ordinary radioactive U-Th halos, they often exhibit outer boundaries that do not match the contour of the inclusion itself. At first thought it would seem the most probable explanation for these halos might be the diffusion of some pigmenting agent from the inclusion into the surrounding matrix. This would raise the possibility that GH may have also formed in this fashion. However, the mass scans in Fig. 3a-c, which show no elemental differences in three separate regions close to and within a GH, do not support this hypothesis. (Admittedly one could say the amount of the pigment was below detection limits so that we cannot completely rule out this hypothesis on the basis of data contained in Fig. 3a-c.) It is nevertheless not clear just how a diffusion phenomenon alone would account for all the observations on the GH. What then is the origin of these GH? While we do not have the final answer, we have acquired some additional information that suggests their existence in these micas may be related to the phenomenon which produced the coloration reversal, for which we now present a working hypothesis.

Formulation of a Working Hypothesis for the Origin of the Giant Halos

First, this Madagascan biotite, as well as all other biotites studied thus far, became darker when irradiated with ^{4}He ions or protons, which is in contrast to the lighter coloration of the halos. It was earlier thought this coloration reversal might have proceeded very gradually over the time period since the biotite crystallized. More recently, however, I have seen U-Th halos with monazite inclusions in what is apparently the same type of mica from the same locale, but with darker coloration than the surrounding mica. This observation made it clear that the long term self-reversal idea was invalid; clearly there was some other factor involved in the coloration reversal. Further investigation showed the only difference in the two specimens was that the mica specimen containing the darker halos was taken from the interior of a larger piece of composite rock whereas the lighter halos were found in separate smaller mica specimens (∿1 cm × 4 cm × 3 cm). This clue would of itself possibly failed to have any significance except for the following information.

Briefly, it is now known that these GH containing micas were first uncovered over twenty years ago during a road cutting operation in the Manangotry Gap region of southeastern Madagascar. It is also known that several years elapsed before H. de la Roche (National Center for Scientific Research, Nancy, France) collected these samples in conjunction with his study (17) of the rocks of that region. During this time the smaller mica specimens were doubtless exposed to various ground water solutions whereas the micas enclosed within larger rocks may very well have remained untouched by these fluids. We therefore advance the hypothesis that the coloration reversal of the halos in this mica was produced primarily by the geochemical interaction of the ground water fluids with the radiation damaged (more chemically reactive) halo regions within the mica.

How does this hypothesis affect the possible origin of the GH? In my opinion we must investigate the possibility that the chemical changes which produced coloration reversal of the halos might also have somehow enlarged their peripheries. Indeed, in this reexamination I have found one halo with a periphery that seems it could have been irregularly altered in just this fashion. On the other hand it is also possible that the chemical changes which produced coloration reversal somehow sensitized the mica, thereby permitting some areas to develop as bleached halos which perhaps were not visible at all before the geochemical interaction with the ground water fluids. In my opinion then we cannot presently exclude the possibility that high energy αs of low abundance may have formed chemically reactive regions outside the

U-Th halos, and further that these reactive regions did not become visible as a halo until after the geochemical interaction with the ground water fluids occurred.

Thus my present position is that we should admit the possibility that the halos in these particular Madagascan mica samples may have more than one mode of origin, and that it may be asking too much to expect that a single explanation can account for all the diverse sizes and apparent stages of development that are characteristic of all the halos in these specimens. Possibly one or more mechanisms acting in concert or sequentially may be operable, and that in spite of the fact that there are many halos which have formed from U and Th α-emitters, we may not be able to necessarily ascribe all halos in these specimens to a first order radiation damage effect produced by α-activity. As noted earlier there are some regular size halos with inclusions that contain little U and Th, which raises the question of whether α-radioactivity was responsible for their existence. Further some and perhaps many GH, especially those with irregularly shaped peripheries, may simply be U-Th halos that were enlarged by the same geochemical interaction that caused the coloration reversal (E. R. Vance, Georgia State University, Atlanta, is investigating the nature of the halo coloration). Yet on the other hand there are GH which appear identical to the U-Th halos except for size. Especially as the outer boundary of some GH matches the contour of the inclusion does the halo appear to be characteristic of a genuine radioactive effect. Although I cannot prove it, to me these particular GH are suggestive of an origin associated with high energy αs.

Indeed it may well be that, notwithstanding the fact that we cannot yet (and may not ultimately be able to) prove that GH originated with high energy αs, the GH did provide enough evidence to cause a search for such activity to be made in Madagascan monazite from the Col du Manangotry region. The first results (14) of that search have provided some evidence of high energy αs. After this α-activity is confirmed and the source identified we will be better able to judge whether it is in any way associated with the origin of the GH found in micas from this area. A GH found (18) in mica from Ireland was actually due to U-Th αs.

Final Results on GH in the Madagascan Mica from the Col du Manangotry Region

The high α-energy isomer hypothesis was also explored, on the assumption that such isomers would eventually decay to Pb, by examining the Pb isotope ratios of the GH and U-Th inclusions with IMMA techniques. This comparison was hindered somewhat because of occasional molecular ion interferences in the mass 200-210 region. In some cases where the mass spectra appeared relatively clear in this region, a few GH and U-Th inclusions showed approximately the same ^{206}Pb/^{207}Pb ratios within a large experimental error (±15%).

While these IMMA studies did not confirm the isomer hypothesis, they did reveal that the group of halos, both GH and ordinary size, which possess unusual opaque inclusions (which are not monazite and presently remain unidentified mineralogically) exhibit a marked and presently unexplained deficiency of ^{208}Pb compared to the monazite inclusions. Compared to the mica (see Fig. 3) they are depleted in Na and K and enriched in Ca, Ti, and Fe (slightly). They also contain what appears to be the same sequence of rare earths as the monazites, but in greatly reduced abundance. Compare for example the IMMA mass scans of the opaque and monazite inclusions shown in Figs. 11-13 with Figs. 3a-c.

One possible explanation of the ^{208}Pb deficiency is that ^{208}Pb has diffused away from these opaque inclusions, but this is extremely difficult to believe because the ^{206}Pb is still present. Another possiblity is that ^{232}Th has only recently been introduced, but the relative constant abundance of ^{232}Th in the opaque inclusion makes this hypothesis equally difficult to believe. Further the IMMA mass scans show no trace of Th in the mica around the inclusions. Another alternative is that the ^{232}Th has accumulated from ^{244}Pu or ^{236}U decay. However, since we did not detect an abnormal number of fission tracks around a few opaque inclusions after they were etched with HF, we know the ^{244}Pu hypothesis is of very minimal probability. The ^{236}U hypothesis does not appear very attractive either because it is very difficult to conceive how or why some ^{236}U would have been selectively incorporated into the opaque inclusions along with ^{238}U and ^{235}U while being excluded from the U composition in the monazite inclusions. Investigations of this very puzzling isotopic anomaly are continuing.

The high mass peaks shown in the IMMA mass scans in Figs. 11-13 are generally attributable to molecular ion combinations of Th, U, Ca, O, and the rare earths. However, the peak at 291 is of interest because spark source studies of this monazite have shown two lines at 145.5.

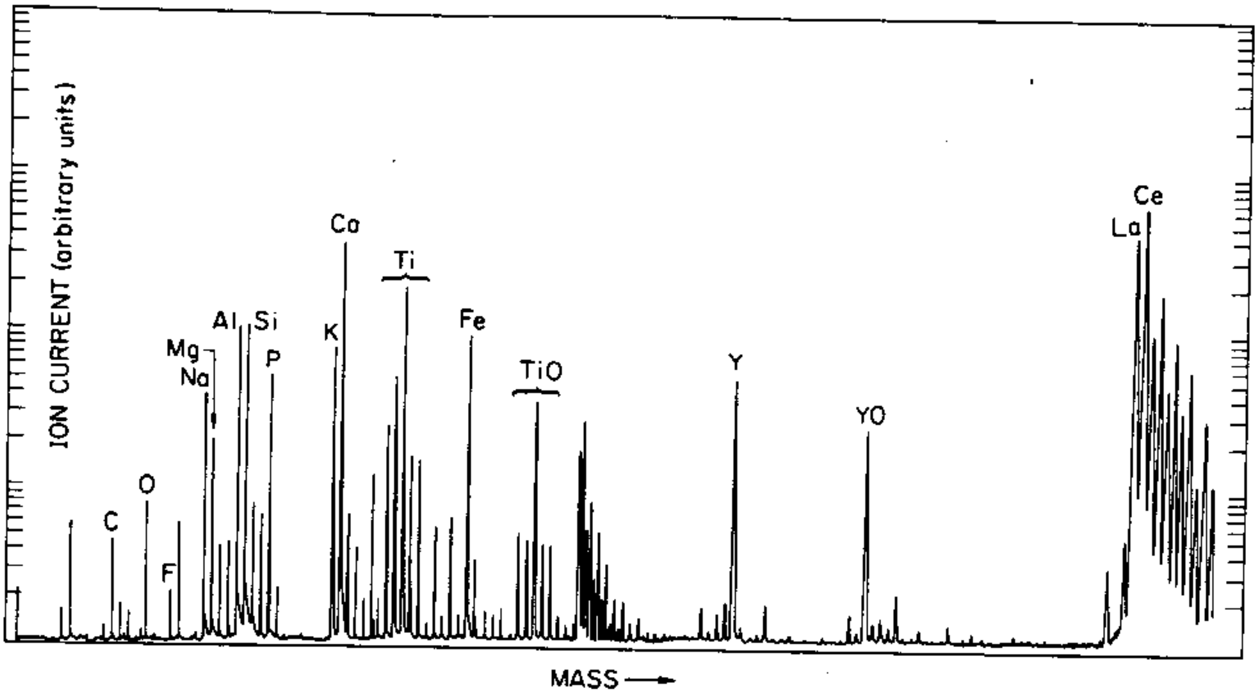


Fig. 11 (a). Mass scan of GH monazite inclusion from mass 1 to ∿150

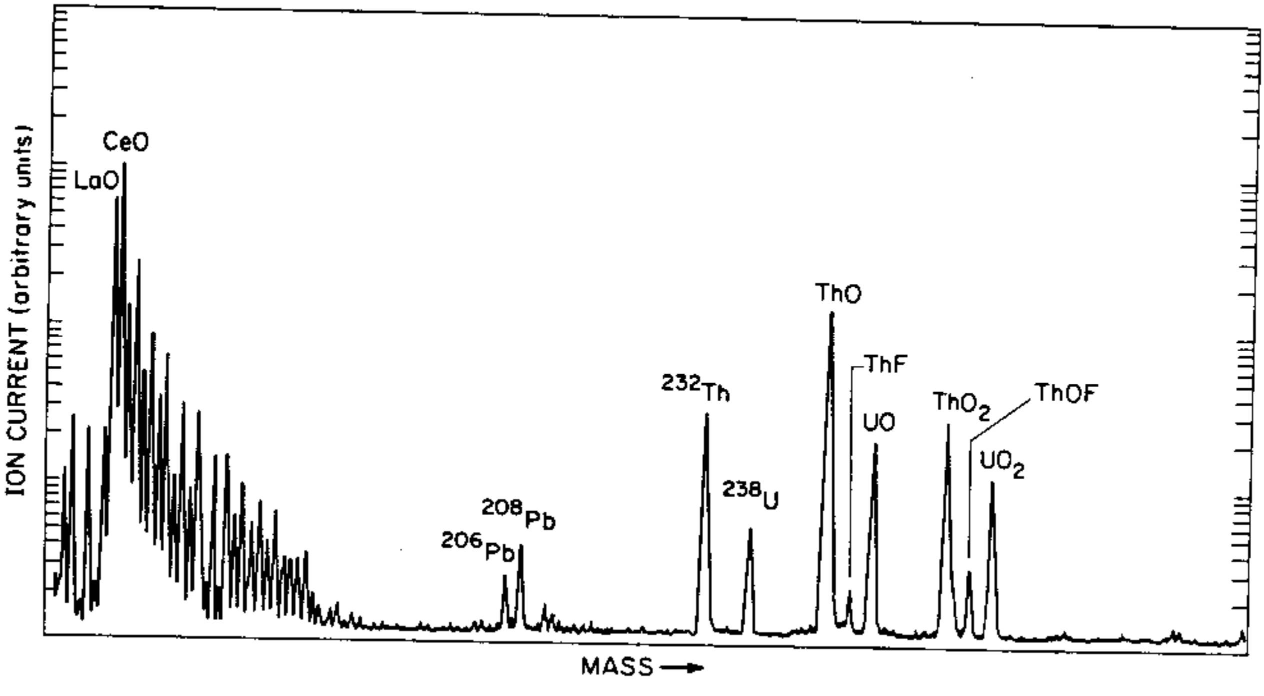


Fig. 11 (b). Mass scan of GH monazite inclusion from mass ∿150 to ∿300

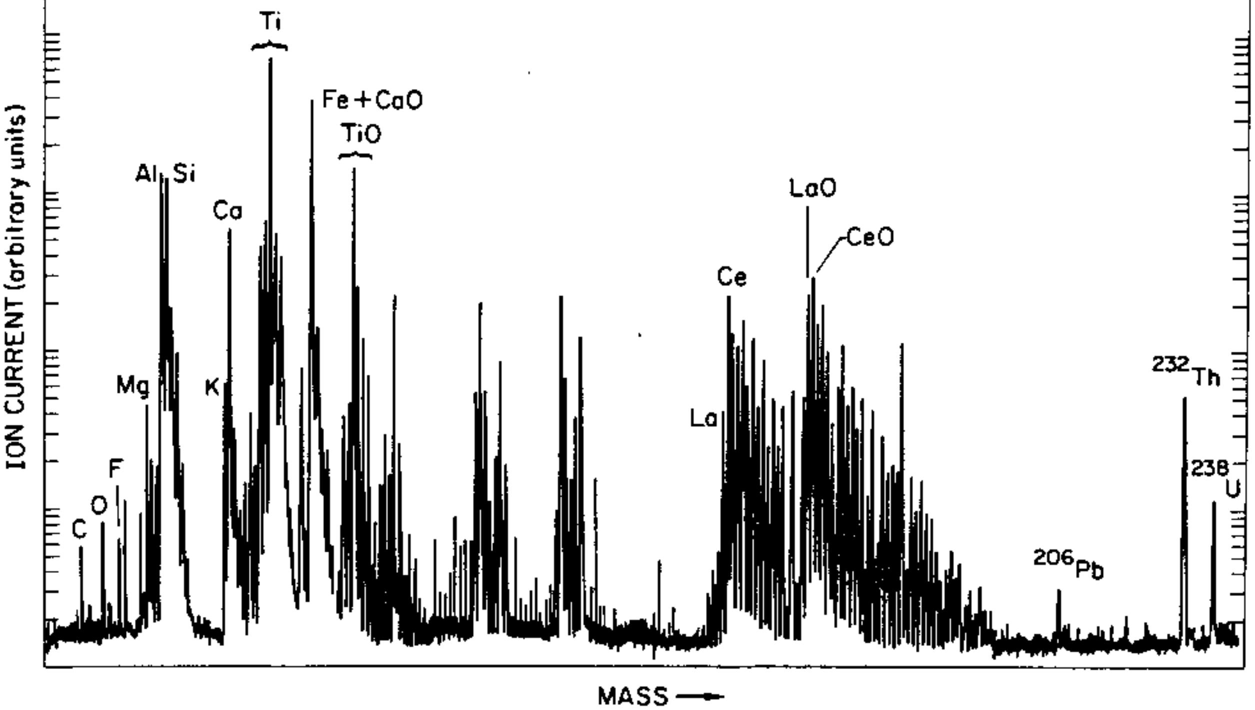


Fig. 12 (a). Mass scan of GH opaque inclusion from mass ∿3 to ∿240

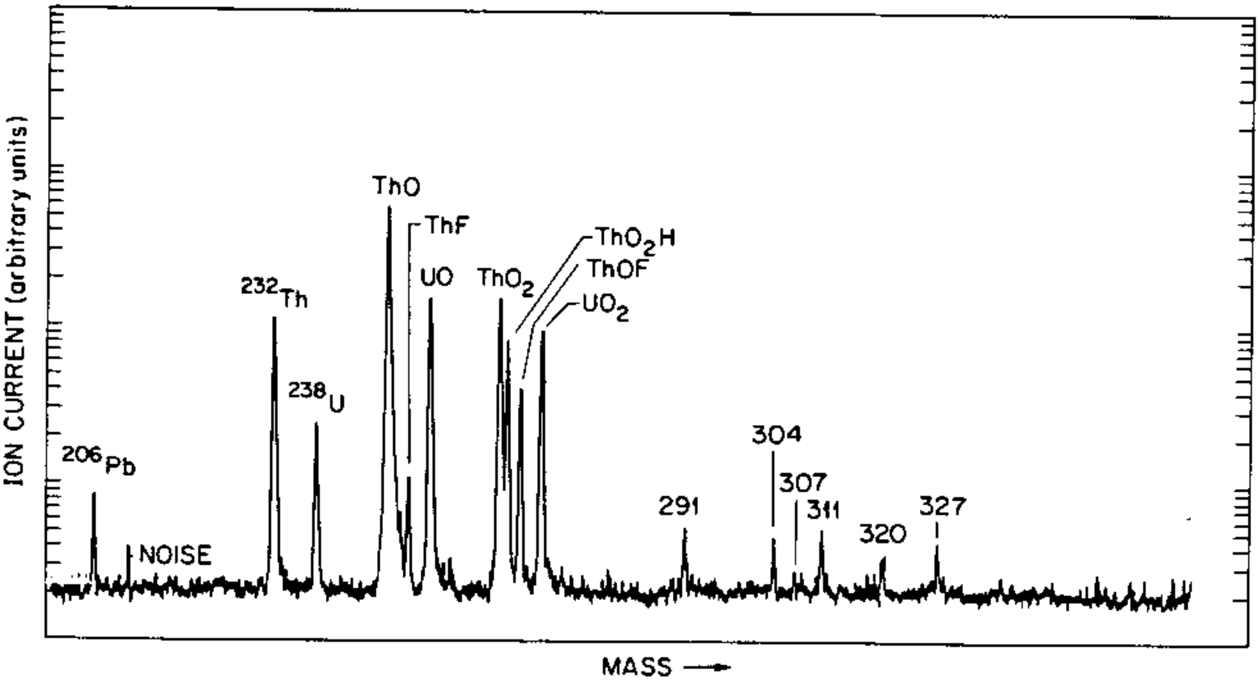


Fig. 12 (b). Mass scan of GH opaque inclusion from mass ∿200 to ∿350

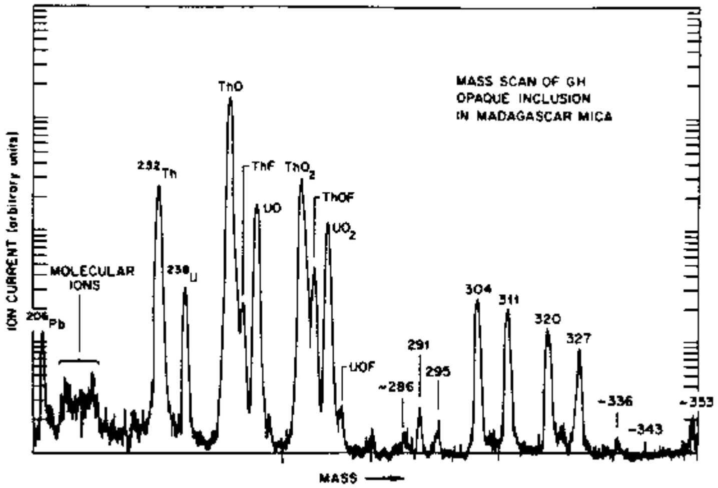


Fig. 13. Mass scan of GH opaque inclusion in Madagascan mica from mass ∿205 to ∿353

Additional experiments are planned so that the masses will be separated (either by a spark source instrument or a mass separator--in collaboration with R. L. Mlekodaj, UNISOR-ORAU) on a foil instead of a photographic plate. This will then allow the mass position at 291 and other high masses to again be analyzed by mass spectrometric techniques using the highly sensitive ion microprobe. If the high masses are molecular ions, they will not reappear as such in the IMMA mass analysis. Neutron activation methods will also be used to study the high-mass-foils.

POLONIUM HALOS

We are also planning on using IMMA, SEMXRF, and PIXE methods to further explore the inclusions of Po halos, both those in rocks (1,2) and in coalified wood (19), for the presence of unusual masses and x-ray peaks. Thus far the research on Po halos has involved mainly analyses which were relevant to the existence of ^{206}Pb from Po decay. The rationale for these projected studies on Po halos stems from the report by Wolke (20), who suggested that element 116, if it exists, might be expected to accumulate in certain marine organisms which are known to have accumulated extremely high concentrations of ^{210}Po. I suggest if indeed element 116 is (or was) primordial and is chemically similar to Po, then there is no place where it would have accumulated in any higher concentrations than in the inclusions of Po halos (1,2,19).

GIANT HALOS IN THE SCANDANAVIAN MICAS

Although giant halos (GH have been reported in Swedish granitic biotite (5,21), Indian pegmatitic cordierites (22), and Madagascan charnockitic mica (5), interest in the phenomenon has centered on the Madagascan GH because of the earlier report (6) implying a possible connection with SHE. Even though this association has not been confirmed (7,8), this renewed interest served as an impetus to re-investigate the GH found in the Swedish biotites, which previously received only minimal attention (5,21) compared to the GH found in the Madagascan micas.

It was found that the great majority of halos in this Swedish biotite are U and Th halos of normal size, the maximum radius being ∿38-40 μm for the Th halo. In about ∿1:1000 halos, however, an inner bleached region which varies from ∿2 to 25 μm in radius surrounds a highly radioactive inclusion. Observations thus far show that when the size of the bleached region is small ($\lesssim$6-8 μm), no change is evident in the dimensions of the halo. However, in those halos in which the bleached region is more intense and of larger radius ($\gtrsim$15 μm), there is generally observed a somewhat weakly colored diffuse outer ring whose radius exceeds the normal dimensions of the halo.

These are the GH which, because they were earlier reported (5) to surround only dense Th halos, were tentatively attributed to the low abundance, high energy αs from ^{212}Po in the ^{232}Th series. Since, however, we now report that diffuse, abnormally large rings also surround dense U halos in this biotite, and since there are no high energy αs of any significant abundance in the ^{238}U chain, it is clear that the original hypothesis is untenable. As an alternative hypothesis we are investigating the possibility that the correlation of the larger-size bleached interior regions with these GH may be a clue to understanding the formation of these GH.

This re-investigation has made it clear that even though these GH were found in Swedish granites obtained from the same locales (i.e., from Rickaby and Arnö) as the specimens Wiman used, the GH described herein are different from the ones Wiman (21) reported. These GH surround radioactive inclusions that are high in U and/or Th (possibly uraninite, thorite, or uranothorite), and have rather diffuse outer boundaries which may vary from ∿42 to ∿55 μm in radius. In contrast, Wiman (21) reported GH in biotite around zircon inclusions showing normal size inner rings and somewhat weak but rather sharply defined outer rings of 57 μm and more rarely of 67 μm. Aside from noting this difference we do not herein comment further on the GH found by Wiman.

To illustrate the unusual characteristics of the GH found in Swedish granitic biotites from Arnö and Rickaby (near Stockholm), we present in Fig. 14 the combined results of applying optical microscopic, electron microprobe x-ray fluorescence (EMXRF) and Ion Microprobe Mass Spectrometer (IMMA) techniques. In particular Fig. 14 shows: (i) a transmitted light optical photomicrograph of a single GH of radius ∿47 μm, (ii) two secondary ion maps obtained (with the IMMA) by rastering the halo region with a finely focused $^{16}O^-$ beam and collecting the sputtered secondary ion signal at mass to charge ratios of 39 (^{39}K) and 40 (^{40}Ca), (iii) two x-ray maps obtained (using EMXRF) by rastering the same region with a 30 kV electron beam and collecting in sequence the K Kα and Ca Kα x-rays, and (iv) the complete EMXRF x-ray spectra obtained by spot focusing the electron microprobe beam first on the mica completely outside the GH and then on the bright circular area within the GH. In Fig. 14 note that the secondary ion and x-ray maps, as well as the contrasting x-ray spectra, show the region corresponding to the bright circular area in the optical photo (which differs slightly in magnification from the maps) is considerably diminished in K (by about a factor of 10) and slightly enhanced in Ca compared to the surrounding mica. Also note that the secondary ion and x-ray maps in Fig. 14 are reflective of the composition of the halo region several micrometers *above* the central radioactive inclusion, the rectangular outline of which can be seen in the optical photo in Fig. 14. Certain of the mineralogical aspects relating to this phenomenon have been reported previously by Rimsaite (23). Fig. 15 shows similar data for another GH in this Swedish mica.

Since the radii of the bright circular areas in the halos in Figs. 14 and 15 approximate the range of the predominant lower energy αs of the U and Th series, we suggest as a working hypothesis the idea that extreme radiation damage effects may have first produced a partial decomposition of the mica in this region with secondary effects then inducing the migration of K out of and Ca into this region. Indeed, the depletion of K has been so great (see Figs. 14 and 15) that backscattered electron maps of the K-Ca inversion region showed a different albedo (see Fig. 15) than the surrounding mica and even the reflected light microscope reveals a coarse, granular surface for this region. Fig. 16 shows the EMXRF spectrum typical of a GH inclusion which is rich in U. Other GH inclusions show high U and Th content.

Admittedly as yet it is not known whether the existence of this K/Ca inversion region is related to the formation of these GH. But, again as a working hypothesis, we must explore the question of whether the present depletion in K may have been accompanied by depletion of other major elements during past epochs. If so, then perhaps for a certain period of time the total mass within this region was reduced sufficiently to allow the highest normal energy αs of the Th and U series (8.78 MeV and 7.68 MeV respectively) to penetrate beyond the normal halo boundary because of having first passed through a region of lower density. Of course, due to the large K depletion and slight Ca enrichment, this region is even now of slightly lower density than the adjacent mica. But unless O, which has not as yet been measured, is also presently depleted, the K (comprising only ∿5-10 atomic % of the biotite) depletion alone would not be sufficient to permit normal energy αs to gain enhanced penetration of ∿15 μm beyond the periphery of a normal U-Th halo. It is certain these GH did not result from diffusion of radioactivity into the mica because IMMA studies showed U, Th, and Pb were confined to the inclusion.

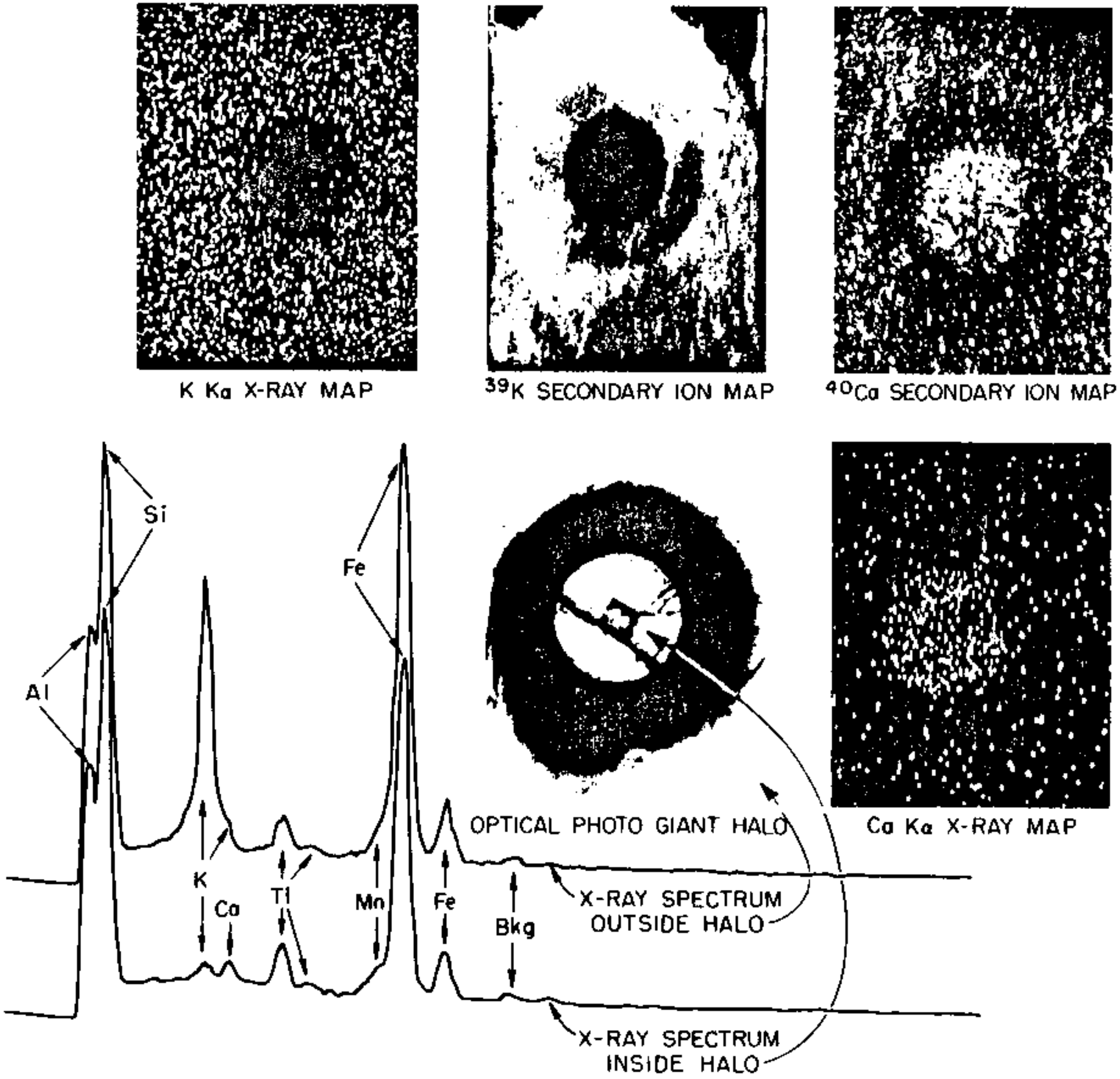
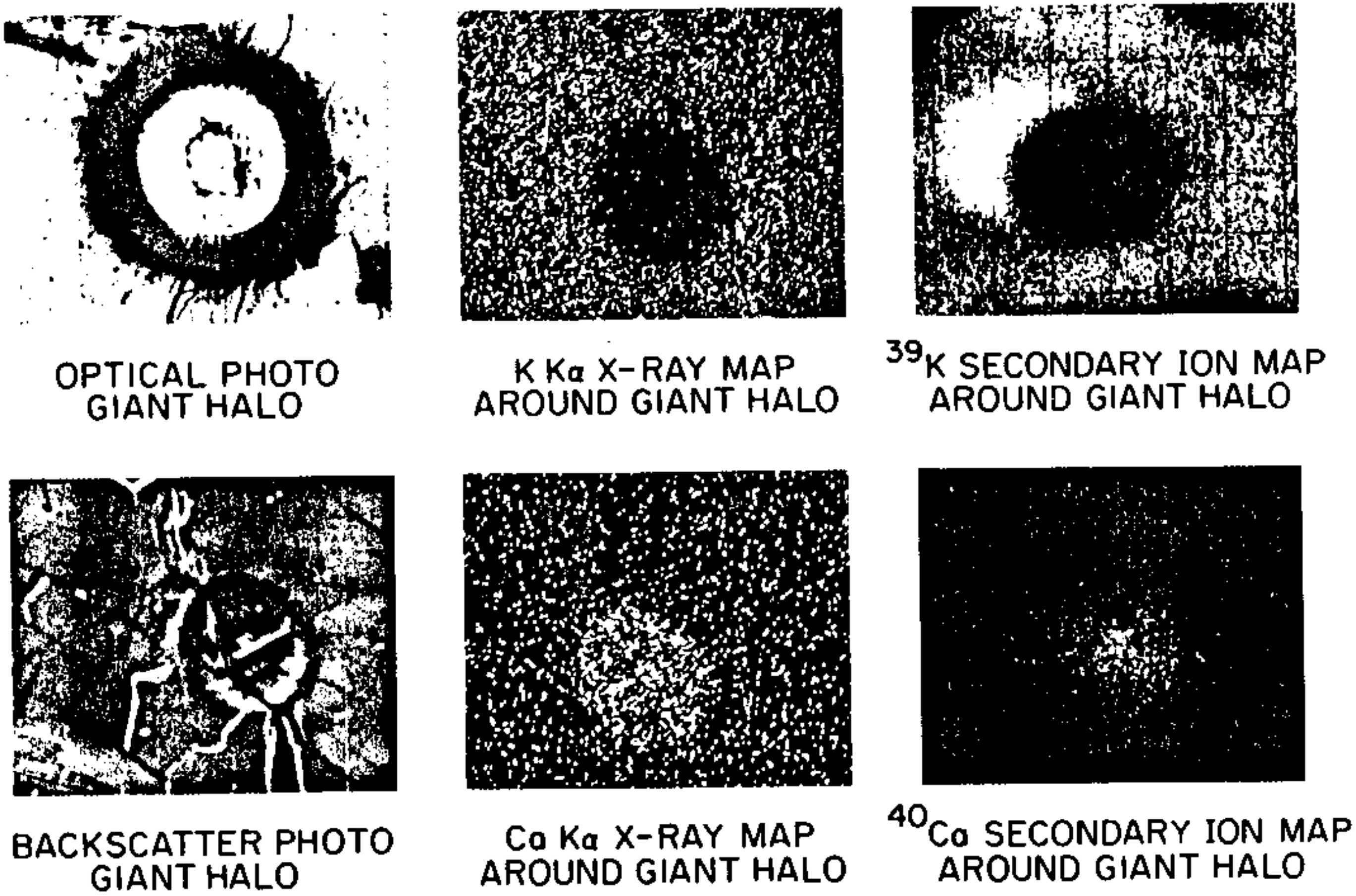


Fig. 14. Optical microscopic, electron microprobe and ion microprobe study of a GH in Swedish biotite



OPTICAL PHOTO GIANT HALO

K Kα X-RAY MAP AROUND GIANT HALO

^{39}K SECONDARY ION MAP AROUND GIANT HALO

BACKSCATTER PHOTO GIANT HALO

Ca Kα X-RAY MAP AROUND GIANT HALO

^{40}Ca SECONDARY ION MAP AROUND GIANT HALO

Fig. 15. Ion microprobe and electron microprobe studies of a giant halo in Swedish granitic mica

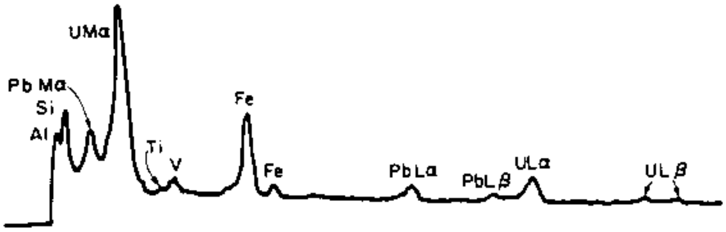


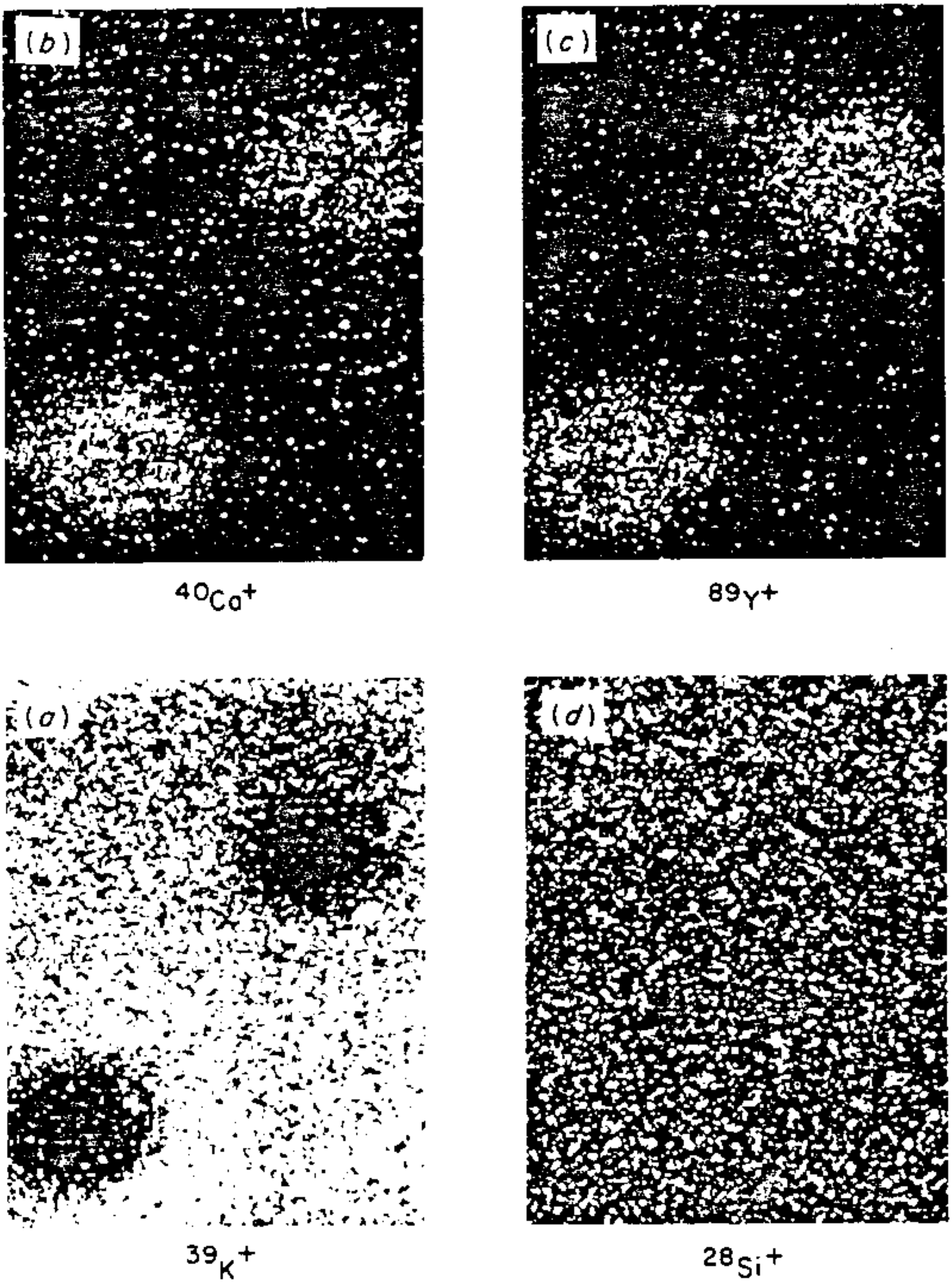
Fig. 16. X-ray spectrum typical of giant halo inclusion in Swedish mica

It appears certain that, irrespective of how the bleached area relates to the GH, it is associated with a highly radiation damaged region. This has had a very important application to studies on the dwarf halos (24,25). In analyzing the dwarf halo centers for the presence of some recognizable parent and/or daughter radionuclides, it became apparent that the dwarf halos exhibited the same type of K-Ca inversion phenomenon as was observed in Figs. 14 and 15. For example, Fig. 17a-d shows several secondary ion maps obtained as an $^{16}O^-$ beam was rastered across two closely spaced dwarf halos. The IMMA ion maps in Fig. 17a-d reveal that these dwarf halos were highly depleted in ^{39}K and enriched in ^{40}Ca and ^{89}Y with no change in ^{28}Si. The complete mass scans shown in Fig. 18a,b, contrasting the dwarf halo region (Fig. 18a) with the surrounding mica (Fig. 18b), reveal a corresponding depletion of ^{41}K, ^{85}Rb, and ^{87}Rb. See also the mass spectra in Figs. 19 and 20. SEMXRF x-ray maps of other dwarf halos also showed the depletion of K and enrichment of Ca throughout the entire extent of the dwarf halo, thus showing the IMMA results were not an artifact of the sputtering process. We consider the association of this K-Ca inversion phenomena with the dwarf halos as reasonable evidence of radiation damage, and hence for a radioactive origin of the dwarf halos.

Optical microscopic and SEM studies on etched and unetched dwarf halo specimens also provided some evidence that the dwarf halos were highly damaged regions. Fig. 21 shows a group of dwarf halos in transmitted light and Figs. 22 and 23 show SEM photographs (at different magnifications) of the surface of a mica specimen which intersects a group of dwarf halos. Note that only holes rather than inclusions appear in the center of the dwarf halos on the SEM photographs (Figs. 22 and 23). While the exact reason for these missing inclusions is not presently known, we could speculate that perhaps some unknown radionuclide(s) in the dwarf halo inclusions decayed to a relatively volatile element. Alternatively we could also speculate that the original radionuclides may have decayed primarily in a fission mode which resulted in the emission of more than two fragments.

HF etching of specimens containing dwarf halos has proved especially interesing. Fig. 24, which is a reflected light photomicrograph of several dwarf halos, shows the halo rim has been preferentially etched compared to the the inner region of the halo. Figs. 25 and 26 show SEM photographs of other etched dwarf halos taken at rather low and rather high magnifications, and these photographs also show the halo rim to be preferentially etched. This more rapid etching of the halo periphery suggests the emission of a particle from the halo center that caused greater damage (higher specific ionization) near the end of its path. While this is a characteristic of α-particles, except for minute amounts of U found in some IMMA scans, we have as yet failed to find evidence that ^{147}Sm (26) or any other low energy rare earth α-emitters produced the dwarf halos, i.e., no significant concentrations of these nuclides were seen in IMMA mass scans of the halo center. Instead we found that several rare earths were uniformly enriched (Fig. 18a) throughout the halo, compared to the surrounding mica (Fig. 18b). See also Figs. 19 and 20. Even though Fig. 18b is devoid of rare earths, other IMMA and spark source mass studies showed these elements do exist in the mica in very low abundance. We tentatively suggest that mechanism for this enrichment is the same which caused the preferential transport of Ca (Fig. 17b) into the halo region (the ion maps of Ca and the rare earths are equal in extent).

In summary, despite this new evidence for a radioactive origin of the dwarf halos, we still cannot explain them on the basis of known low energy α-activity. Despite also the fact that



$^{40}Ca^+$ $^{89}Y^+$

$^{39}K^+$ $^{28}Si^+$

Fig. 17. Four different secondary ion maps of an area in the Ytterby mica containing two dwarf halos

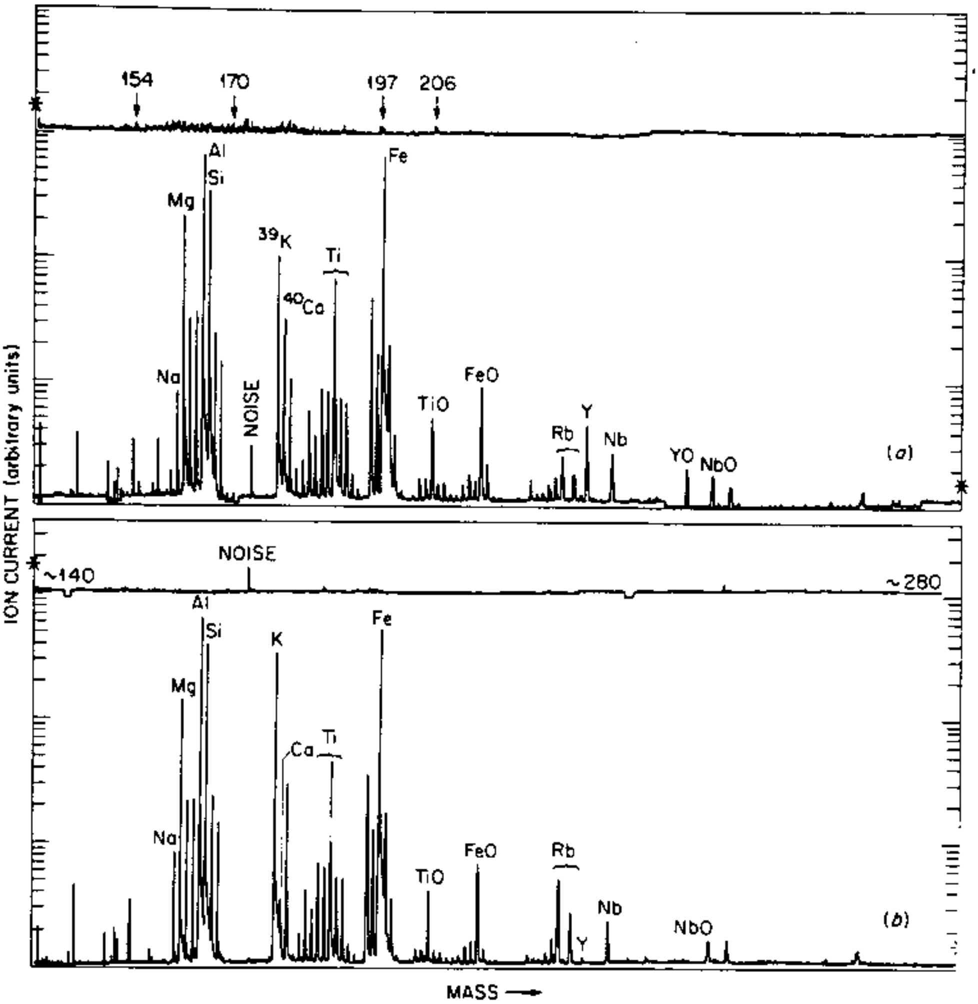


Fig. 18. (a) Mass scan of dwarf halo. (b) Mass scan of mica adjacent to dwarf halo.

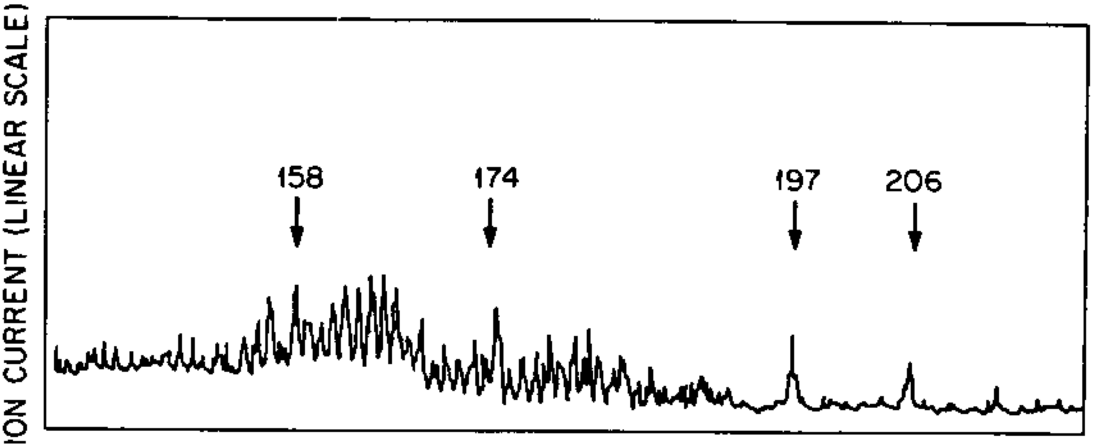


Fig. 19. Mass scan of dwarf halo

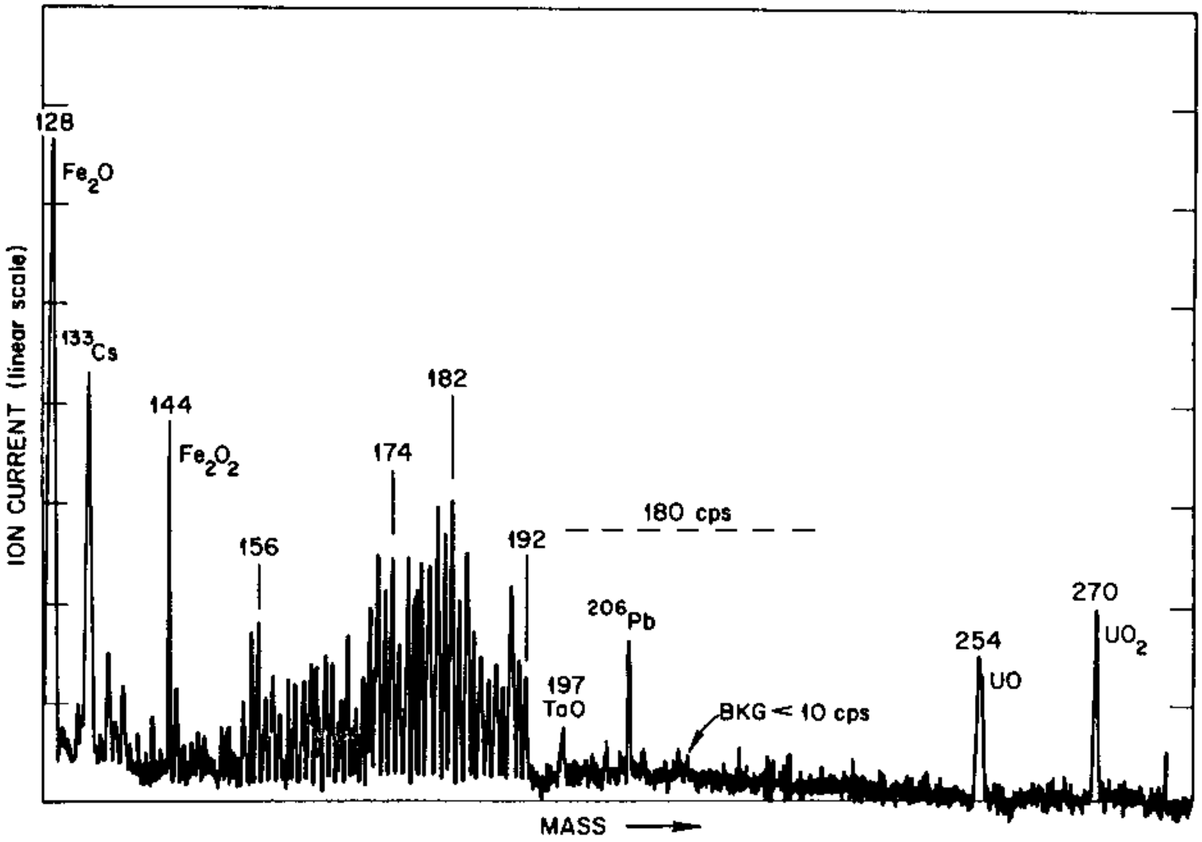


Fig. 20. Mass scan of dwarf halo in Ytterby mica

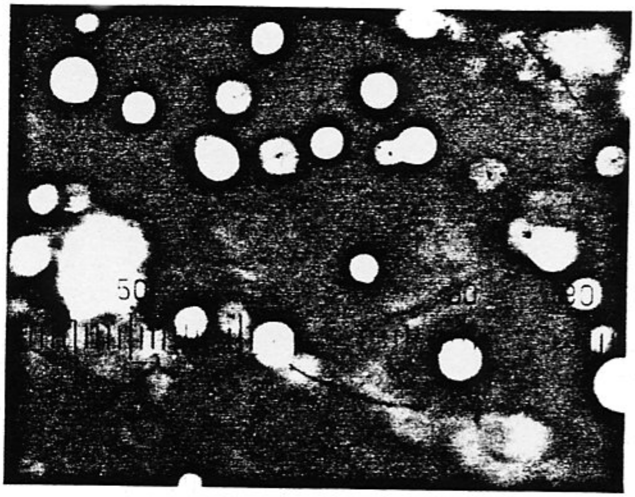


Fig. 21. Optical photomicrograph of a group of dwarf halos in Ytterby mica. Each scale division ≈ 2.5 μm.

Fig. 22. Scanning electron microscope photograph of a group of unetched dwarf halos. Aprroximate magnification ∿500.

Fig. 23. Scanning electron microscope photograph of a group of unetched dwarf halos. Approximate magnification ∿900.

Fig. 24. Reflected light optical photomicrograph of a few etched dwarf halos. Approximate magnification ∿850.

Fig. 25. Scanning electron microscope photograph of a group of etched dwarf halos. Approximate magnification ∿270.

Fig. 26. Scanning electron microscope photograph of a single etched dwarf halo. Approximate magnification ∿2000.

unknown low energy αs have been reported by Brukl et al. (27), we are not now inclined to associate the dwarf halos with this α-activity because out of the thousands of dwarf halos examined, none exhibit the dark coloration characteristic of normal halos in this mica. We therefore speculate that dwarf halos were produced by ions heavier than αs but with shorter range than normal fission fragments. The much higher specific ionization of such particles would be expected to produce coloration and reversal effects at far lower doses than that required with α-particles. Such reduced quantities of the parent nuclide might result in quantities below our present detection limits. In this respect Carvalho et al. (28) report the possible discovery of a short range heavy ion decay mode in U impregnated nuclear emulsions. When more data on this phenomenon becomes available we will be able to assess whether it is in any way associated with the dwarf halos.

Conclusions

Evidence has been presented showing that the dwarf halos formed from a presently unknown type of radioactivity. The question of whether the dwarf halos or X halos originated with radioactivity in the SHE region is presently under investigation.

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DISCUSSION

R. Lindsay: I realize that there are many possible explanations for the giant halos the one that we investigated was α-p reactions in aluminum, silicon, potassium and other elements which are on mass scans. We detected about one proton. Now the protons are fairly sharp at their upper limits for every 100,000 elements I wonder if you will comment.

R. V. Gentry: We can estimate a minimum of 10^9 protons would be needed to produce a GH from (α,p) reactions on low Z elements because it requires about 10^9 α,s to produce a normal U-Th halo: The extended volume in which the (α,p) reactions occur could possibly increase this number to 10^{10} or $10^{11}$for threshhold coloration from protons. The problem is that in giant halos with small inclusions perhaps not more than 10^{10} to 11^{11} α,s have been emitted from the U and Th α-emitters contained in these inclusions. Thus, if only 1 in 10^4 or 10^5 α-interactions with low Z elements results in proton emission, then we are clearly several orders of magnitude below the necessary proton fluence needed for coloration at distances corresponding to GH radii.

E. Fireman: We have observed rapid diffusion of Thoron, ^{220}Rn, from monazite and no alpha particles >10.6 MeV. Can't the diffusion of thoron into cracks in the mica and along the crystal plane of mica explain the giant halos?

R. V. Gentry: I think you are talking about ^{220}Rn. Have you looked up the diffusion coefficient for radon in monazite?

E. Foreman: Well, I have seen it come out of both samples very rapidly and I see sharp lines corresponding to Th and I see the daughter deposited on the face of the crystal. I pull the sample away with the 10.7 activity. I see the daughter decay on the face of the crystal at 10.7 MeV above the sample and there is quite a bit of activity.

R. V. Gentry: Well, let me continue. The diffusion coefficient for radon in natural crystals is very low ($<10^{-22}$ cm^2/sec). Now the radon isotope you are referring to has only a 56 sec half-life.

E. Fireman: 55 not 56. That is what surprised me. It is out there and you see strong lines it diffuses out very rapidly.

Unidentified Speaker: You get along way in 55 seconds. I don't argue about the diffusion qualities. It means very much because that would be something perfectly intact.

R. V. Gentry: The relatively shoft half-life of ^{220}Rn means that on the average it will diffuse only very small r.m.s. distances ($\sim 10^{-5}$ cm) in the crystal before decay. Some radon escapes from the surface via recoil action, but this also moves only small distances unless there is a fissure in the mica. I believe your other question was in reference to our search for lead on the mica surfaces adjacent to the GH inclusions. Sometimes we observe a section 50 μm thick. If I understand what you are saying correctly the radon must diffuse out of the monazite into the 50 μm section. We have examined sections of several GH without seeing any evidence of ^{208}Pb, the decay produce of ^{220}Rn, on the surface of the mica adjacent to the inclusion.

E. Fireman: Just a second, you say 50 micrones. Just test the diffuseness out from the surface of the monazite. I go into the surrounding region and get out micron a certain distance 20 microns or so and then you get the result the 8.6 daughter, 8.6 MeV and 10.6 MeV daughter of the radon 220. Those alpha arise from 20 microns from the inclusion or 25 microns.

R. V. Gentry: Well, you would with the ion probe.