Reports

Polonium Radiohalos: An Alternate Interpretation

Abstract. *A study of the sizes of so-called polonium radiohalos of various types found in biotite from Bancroft, Ontario, has been carried out. The evidence is consistent with the interpretation that these halos are variants of the standard uranium halos. A review of the literature indicates that there is no firm evidence that polonium halos exist, all evidence being equally consistent with the interpretation that these are uranium halos.*

It has been more than 30 years since Henderson and Sparks (*1*) first suggested the existence of polonium radiohalos (*2*). These are one-, two-, or three-ringed structures created by the alpha decay of ^{218}Po, ^{214}Po, and ^{210}Po but lacking the rings due to the other five alpha decays from the ^{238}U series. The independent existence of polonium halos, distinct from their being variants of the standard uranium halo, is still in doubt. We question a number of points in recent work by Gentry (*3, 4*) concerned with the determination of the true nature of these halos.

We now report the results of a series of measurements made on polonium-type halos. Our measurements do not support the polonium halo hypothesis. We cannot definitely rule out the existence of polonium halos, but it appears that there is no evidence requiring, or even firmly suggesting, their existence. It was realized very early (*5*) that their existence would cause apparently insuperable geological problems since the relevant polonium half-life is of the order of minutes. Polonium halos would require that the polonium atoms become part of the inclusion within minutes of the formation of the polonium and that in this very short time the polonium must be so far removed from the parent uranium mass that its presence or location is no longer evident.

Several kilograms of biotite were collected from the Faraday pegmatite at Bancroft, Ontario, Canada. The age of this body is generally accepted to be approximately 10^9 years (*6*). Specimens were taken from several crystals, cleaved to a thickness of 15 to 30 μm, and mounted on petrographic slides. Radiohalos, all of the so-called polonium types, were relatively abundant, averaging about one per square centimeter. Halos exhibiting rings attributable to alpha decay from other members of the ^{238}U series were not observed, nor were any thorium halos or halos of any other type. The number of halos observed was considerably greater than the number that were measurable.

According to the classification of Henderson and Sparks (*1*), there are three distinct kinds of polonium halos. Type A is due to the decay of ^{210}Po and consists of a disk only. Type B consists of a disk due to ^{210}Po and an outer ring due to ^{214}Po. (The assignment of the type B disk to ^{210}Po is due to Henderson and Sparks.) The third kind, type C, consists of a disk, an outer ring, and an intermediate ring, slightly larger than the disk, due to ^{210}Po, ^{214}Po, and ^{218}Po, in that order. A fourth type, not previously reported, which we labeled type AB, consists of a disk and a slightly larger ring due to ^{210}Po and ^{218}Po, respectively.

According to Henderson and Sparks, the size of the type B disk is the same as that of the type A or type C disk and comes from the alpha decay of ^{210}Po. Should this be the case then the absence of the intermediate ring (^{218}Po) rules out type B as a uranium halo, for if the larger ^{214}Po ring appears, the smaller ^{218}Po ring must also appear since that radiation would be present in a uranium halo.

However, should the type B disk be the size of the type C intermediate ring (^{218}Po), the halo may be interpreted as a darkened example of a uranium halo where the disk and intermediate ring have blended. Before the reports by Henderson and co-workers (*1, 5, 7, 8*), Joly (*9*) identified and measured three uranium halos which are visually indistinguishable from Henderson's three types. [Compare figures 3 and 4 in plate 1 of Joly (*9*) with figures 5 and 4, respectively, in plate 6 of Henderson and Sparks (*1*). We must point out here that there is a ^{222}Rn ring which comes from an alpha decay of energy just 3.6 percent higher than the ^{210}Po alpha energy. This ^{222}Rn ring and the ^{210}Po one are not resolvable.]

The results of our measurements are given in Table 1. The size of the type B disk is consistent with the size of the intermediate (second) ring of type C and completely inconsistent with the smaller disks of types A, AB, and C. The smallest type B disk we found measured 20.9 ± 0.6 μm, whereas the largest type C disk measured 19.8 ± 0.6 μm. None of the 24 disks of types A, AB, or C had a measured value larger than the smallest type B disk, and the mean value for the type B disk lies more than five standard deviations from the mean of all 24 disks of types A, AB, and C.

Our findings do not agree with those of Henderson and co-workers (*1, 5, 7,*

Table 1. All measurements are of ring radii, in micrometers, with probable errors as given. The number in parentheses after each value is the number of samples measured. For the first ring, the outer part of the second ring, and the third ring, measurements were made to the outer extremity. "Inner" refers to the inner dimension of the second ring and "maximum" indicates the point of maximum darkening of the second ring. The mean value for the second ring for type B is more than five standard deviations from the mean value for all 24 measurements for the first ring. The errors are determined from the estimated probable error of measurement and are not standard deviations from the mean.

Radiohalo type	First ring	Second ring			Third ring
		Inner	Maximum	Outer	
A, disk only	19.9 ± 0.2 (13)				
AB, two inner rings	19.1 ± 0.3 (4)		21.0 ± 0.5 (2)	22.2 ± 0.5 (2)	
B, inner and outer rings				22.3 ± 0.3 (5)	35.5 ± 0.3 (5)
C, three rings	19.3 ± 0.2 (7)	20.9 ± 0.5 (2)	21.8 ± 0.3 (5)	23.1 ± 0.4 (3)	34.8 ± 0.3 (7)

8) for the size of the type B disk. For the outer ring of type B as well as for other rings of all types, our measurements are consistent with theirs.

Despite the fact that our biotite came from a pegmatite considered to be a source of polonium halos, we conclude that the halos we measured are darkened uranium halos. The existence of the previously unreported type AB showing rings from ^{210}Po and ^{218}Po indicates that these halos exist in varying stages of formation. In this case the ^{214}Po alpha decay must be there, and its outer ring is missing (or in some cases, such as in Fig. 1, only very faintly visible) only because the amount of radiation has not been sufficient to darken this ring. The halos may be viewed as progressing sequentially through stages in the following order: A, AB, C, B (see Fig. 2).

In light of Joly's early measurement and identification of various one-, two-, and three-ringed structures as uranium halos, how did Henderson decide that he had discovered new types even though what he saw appeared identical to Joly's types? The apparent answer, that Henderson measured type B disks that were smaller than Joly's, is not supportable. In order to do this, Henderson needed a method to classify a halo as type B before making the measurement, for the distinction between Henderson's type B and type C halos is, by his own report, not reliable.

Henderson and Turnbull (*8*) measured 26 type B halos, but in 9 of these they also saw measurable intermediate rings and in others they saw evidence of the intermediate ring. They remarked on the presence of these intermediate rings in the following way: "It is probable that these rings may be due to halos of type C wrongly classified, for these are sometimes difficult to distinguish from type B." Whereas such halos should obviously not be counted as type B, Henderson and Turnbull so classified them solely on the basis that type C halos had more discernible intermediate rings. Their basis for new halo types was a subjective judgment of the intensity of coloration of various rings. This procedure ignores the fact that different micas show marked differences in response to the same radiation dose (*7, 8, 10, 11*). Differences of as much as a factor of 5 in absorbance have been observed for different micas irradiated with the same alpha dose (*10*). Further, there are different levels of darkness at which different micas saturate, and the curves obtained by plotting darkening against alpha dose vary in shape for different micas and are nonlinear (*10*).

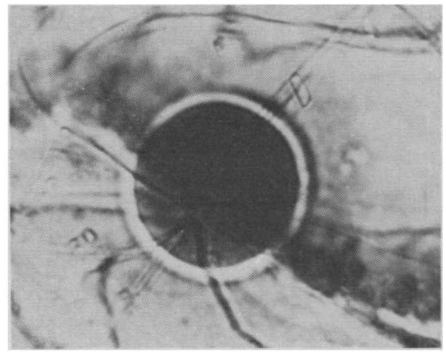

Fig. 1. Type AB radiohalo, showing a ^{210}Po disk, a partly developed ^{218}Po ring, and a very faintly developed ^{214}Po ring. The average radius of the inner disk is 19.1 μm.

The apparent darkness of a halo is also affected by the thickness of the thin section. Factors related to visual perception cannot be discounted either. It is apparent from plate 7 in (*8*) that the appearance of the intermediate ring must be judged against a dark background, which complicates visual interpretation.

Henderson and Turnbull (*8*) noted, but could not explain, that their own polonium ring measurements were systematically higher than their corresponding measurements for these rings in uranium halos. For example, their radius for the ^{218}Po ring was 3.6 percent larger in polonium halos than in uranium halos. Other shortcomings of the work of Henderson and co-workers have been noted in (*12*).

Gentry (*3, 4*) has looked for uranium itself in the polonium inclusions by two means, by using slow neutron fission tracks from ^{235}U and by an examination of the inclusion with the ion microprobe (*13, 14*). Gentry (*4*) concluded that uranium either does not exist or is present only in minute quantities in the inclusions. We believe, however, that there are other interpretations which have been overlooked.

Gentry (*4*) indicated that the fission and ion microprobe techniques do show uranium (at least sometimes) but he considered the amount to be one to three orders of magnitude too low to cause coloration. We consider the natural radioactive flux necessary for coloration (frequently given as 10^{14} particle/cm^2) to be known only to two orders of magnitude, at best because (i) the minimum measurable darkening is poorly defined and (ii) the sample-dependent variation of the minimum flux necessary for darkening can be an order of magnitude. Since Gentry (*4*) reported no direct comparison between the amounts of uranium found in the polonium inclusions and the amounts of uranium in a series of control uranium halos, we are reluctant to accept his conclusions.

Another serious problem with the ion microprobe work is that it is essentially a surface technique with a maximum resolution of 2 μm (*14*). In order to study the composition of a halo inclusion, it would be necessary to ascertain that the inclusion lies on the surface of the thin section and that it is large enough and placed accurately enough within the instrument that the microprobe would not detect a significant portion of the host environment. Since the inclusions are approximately 2 μm in diameter and are generally embedded within the biotite (which is

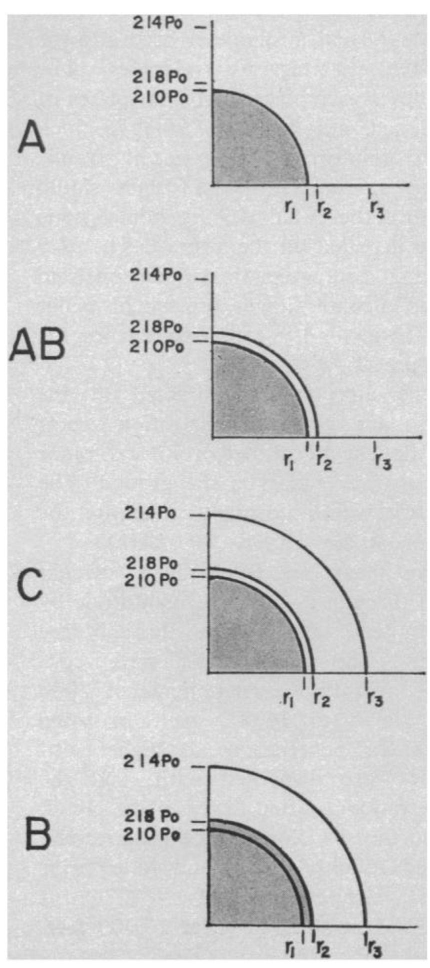

Fig. 2. Schematic sequential diagram of the halo types as described in the text; r_1, r_2, and r_3, are the radii of the first, second, and third rings. The sizes of the three radii can be found in Table 1.

usually 15 to 20 μm thick), and the microprobe must operate at the extreme limits of its capability, it seems likely that the microprobe is examining the host environment along with the inclusion.

Gentry (*4*) reported an anomalously high ^{206}Po/^{207}Pb ratio for one inclusion. This anomalous value may be understood in a number of ways. It can be interpreted as reflecting a supporting ^{238}U/^{235}U ratio higher than the usual value by a factor of 3 or more. (In addition, this would explain the reduced evidence of slow neutron fission tracks since ^{235}U would be underabundant.) While it is well known that this ratio is extremely constant over a variety of rocks, all such determinations apply to macroscopic averages (*15*). No determination of the constancy of this ratio over regions of microscopic dimensions has ever been made. The nearest such measurements are for regions more than nine orders of magnitude larger in volume. There is no reason to believe that on a scale of a few micrometers this ratio should be constant. The fact that two different samples of the same age yielded ^{206}Pb/^{207}Pb ratios differing by a factor of 3 can be interpreted as indicating that the supporting ^{238}U/^{235}U ratio does vary on a microscopic level. It is also apparent that if the microprobe is "seeing" the host as well as the inclusion, then the lead results are misleading.

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15. Both terrestrial and lunar materials have ^{238}U/^{235}U ratios that are determined to five significant figures and are consistent with each other. However, all techniques used in such determinations deal with quantities of material larger than 0.1 g. This corresponds to volumes larger than 0.1 cm in linear dimension. In our case we are interested in volumes of the order of 10^{-4} cm in linear dimension. Thus, the best ratio determinations come from regions at least three orders of magnitude larger (in linear dimension) then the scale relevant to our problem. See P. R. Fields, H. Diamond, D. N. Metta, C. M. Stevens, D. J. Rokop, in *Proceedings of the Second Lunar Science Conference*, A. A. Levinson, Ed. (M.I.T. Press, Cambridge, Mass., 1971), vol. 2, p. 1571; J. N. Rosholt and M. Tatsumoto, in *ibid.*, p. 1577; A. Turkevich, G. W. Reed, Jr., H. R. Heydegger, J. Collister, in *ibid.*, p. 1565.

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Water Content in Convective Storm Clouds

Abstract. *The condensed water content of convective storms was measured by the use of a penetrating aircraft. Regions 1 to 2 kilometers in extent and having condensed water contents of about 20 grams per cubic meter were found to be definite features of the cloud interior.*

Measurements of the condensed water content of severe convective storm clouds were carried out during the summer of 1972 as a part of the operations of the National Hail Research Experiment. These measurements were obtained by the use of an armored aircraft operated by the South Dakota School of Mines and Technology.

The aircraft was a T-28 with the leading edges armored by the addition of heat-treated aluminum 0.090 inch thick. This thickness was found to be sufficient to withstand impacts by balls of ice 7.5 cm in diameter fired into the surfaces at aircraft velocities. The canopy was replaced with flat plates of stretched acrylic 0.60 to 0.75 inch thick reinforced by a metal frame. Since the impact of hailstones would rupture the usual deicing boots, none were installed on the aircraft. The only deicing equipment installed consisted of an alcohol slinger for the propeller and an alcohol injection system for the carburetor.

The aircraft was directed to the storm and given a penetration vector by the use of a meteorological radar and a track radar on the ground. The aircraft would normally penetrate the storm at an altitude of 24,000 feet above mean sea level, fly a straight path through it, decrease its altitude by 2000 feet, and return through the storm. The penetrations were continued at altitude decrements of 2000 feet down to 16,000 feet. In some cases the penetrations at higher altitudes were dispensed with, and the penetrations started lower in the cloud. In northeast Colorado, the penetrations would roughly correspond to exterior cloud temperatures of $-22°C$ at 24,000 feet and $-1°C$ at 16,000 feet. The cloud base varied from day to day, but was approximately at 5°C and 11,000 feet. The surface elevation was 5300 feet above mean sea level.

The condensed water was measured by the use of an electrically heated evaporator mounted below and forward of the wing tip. The evaporation took place in a tube 22 cm long; lined with low-mass heating elements on the inside surface. The inner diameter of the tube was 1.8 cm, but the entrance diameter was 0.8 cm. The limitation to the latter size was dictated by power availability. Six screens, having a 1.4-mm mesh, were spaced along the length of the tube. The water drops that passed through the entrance tube struck the screens and broke up into smaller drops, which were quickly evaporated. A similar instrument has been describd by Ruskin (*1*), who found that individual drops were evaporated in 0.1 second. A thermistor at the exit of the evaporator was used in a control circuit to maintain a constant output air temperature of 90°C. The output temperature and the power supplied to the heating element were recorded on a digital tape recorder each 0.63 second along with the other required parameters of cloud temperature, airspeed, and pressure. The response time of the evaporator was limited to 3 seconds by the temperature control circuit. Feature *A* in Fig. 1 is approximately 10 seconds wide at the base, and thus illustrates that the response time is very near the 3-second limitation of the temperature control circuit.

The condensed water content was determined from the power required to supply the heat of vaporization for the water; this is the power over and above that required to heat the incom-