The Thermal Erasure of Radiohalos in Biotite

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ABSTRACT

Polonium (Po) radiohalos in biotites in some crystalline rocks have been an unresolved enigma, their presence defying conventional explanations for the formation of the biotites and their host rocks via processes involving heat over long timescales. Experiments were therefore conducted to investigate the affect of heat on biotites and their contained Po radiohalos — at temperatures of 250° to 700°C for between one and five hours. Significant changes and damage occurred to many of the biotite samples, with both structural defects and Po radiohalos being variously erased. The results support the contention that neither elevated temperatures and long timescales during biotite and host rock formation and subsequent cooling, nor the later migration of Po via solutions along microcracks into the radiocenters, can account for these Po radiohalos. These biotites and their host rocks may thus have formed very rapidly, possibly even under cold conditions, or else nuclear decay rates were then much slower.

INTRODUCTION

Pleochroic radiohalos are the microscopic zones of colour and concentric rings which form around minute radioactive mineral crystals or inclusions (radiocenters) as the result of alpha particles produced in the uranium (U) and thorium (Th) decay chains of the trace U and Th in the crystals or inclusions. Such radiohalos have been found by many investigators in biotites in granites, pegmatites, veins and dykes,[1] in fluorite and cordierite, and even in diamonds, an example of the latter having been described previously by one of the authors of this study.[2]

Currently accepted genesis theories for the formation of granites, and the biotites within them, include the widely popularized scenario that hot processes, namely crystallization from liquid magmas, are responsible for the formation of these crystalline rocks and thus the biotites which contain the radiohalos. Similarly, pegmatites, veins and dykes are believed to have crystallized from 'late' residual melts, metamorphic segregations or hydrothermal fluids that intruded along fractures in the host or wall rocks, usually other intrusive igneous or metamorphic rocks, but again heat is involved. Diamonds are said to have crystallized below 150 km depth in the lithosphere and subsequently transported into the crust by rapidly ascending kimberlitic or lamproitic magmas. In addition, it is postulated that all these processes continue to operate in much the same way today deep within the earth's crust and lithosphere.

Crystallization is said to be slow, beginning in some instances at temperatures ranging from 1,200 to 2,000°C (Celsius), because such temperatures are required to form the very large bodies of coarse-grained granites and their biotite crystals, which are contained within the enormous batholiths (some as long as 1600 km) seen on earth. Pellant states:

> *'Because of their size and depth, the magma cools very slowly, possibly taking tens of millions of years from initial injection to freezing'.*[3]

If such is the case, these igneous rocks, and even the veins and dykes produced by metamorphic or hydrothermal activity, would have cooled at only a few degrees or even one degree per year, and therefore must have been subjected to higher than ambient temperatures even after hardening for, at the very least, thousands or even tens of thousands of years.

Some of the radiohalos which occur in granitic, pegmatitic and vein/dyke biotites have been produced by isotopes with very short half-lives. It has also been shown that they appear to be parentless with respect to either the U-238 or the U-235 decay chains. In particular, the polonium (Po)-218 radiohalos, identified by Gentry,[4] have been produced by the Po-218 isotope which has a half-life of only three minutes, and have radiocenters which show, via electron microprobe mass spectrometry analyses, that the Po-218 had not been a part of the natural U decay process within those radiocenters, that is, it appears to have been parentless. Gentry has also clearly shown that no gross transport of alpha radioactivity in solution or by way of diffusion of radon (Rn)-222 gas could have been passed through thin clefts or microcracks in the minerals to the radiocenters at the halo formation sites.[5]

In many studies, including this one, radiohalos have been found and photographed in the absence of any apparent microcracks or clefts in the biotite crystals/grains. This implies that either

(1) the Po-218 that was in the inclusions (radiocenters), as well as all the elements that now make up the inclusions and the surrounding biotite matrix, were all present together at nucleogenesis and that very rapid crystallization of the inclusions and biotite followed, or

(2) the radioactive Po-218 migrated in solution, or its gaseous Rn-222 parent diffused, into the radiocenters at the sites of halo formation some time after crystallization of the biotite and its inclusions via microcracks which were later healed, presumably by associated heat.

If these large biotite grains containing the Po-218 radiohalos (see Figure 1) crystallized, for example, from hot granitic melts which took up to millions of years to cool, and the radiohalos formed immediately after such granite crystallization, then these short half-life radiohalos must have been exposed to temperatures in excess of 500°C for a protracted period during the cooling histories of the rocks. Furthermore, even if the Po-218 (or Rn-222) had migrated some time later into the inclusions via microcracks in the biotite crystals which had subsequently healed, any heat associated with the migration process (for example, hydrothermal activity) and/or imposed on the granite, pegmatite or vein/dyke subsequent to their crystallization would have likewise exposed the radiohalos to protracted periods of elevated temperatures.

Fossil fission tracks etched into mineral surfaces, also associated with the decay products of U, have been subjected to thermal stress by many investigators, including Roth and Poty,[6] who have shown that fission tracks in muscovite, another mica mineral, can be erased from the mineral at about 500–600°C in one hour or less. Sandhu and Westgate state:

> *'Heat is by far the most significant environmental factor which causes partial or complete fading of spontaneous (fission) tracks.'*[7]

They go on to point out the work of Naeser *et al.*,[8] who demonstrated that fission tracks in obsidian are annealed from the rock even at ambient surface temperatures over long time periods.

Figure 1. *Photograph of one of the biotite grains with fully developed Po-218 radiohalos in it used in our experiments (sample IM-1, 1X magnification).*

The purpose of this study was to investigate how short term exposure to high temperatures and longer term exposure to a moderate temperature affects Po-218 (and Po-210) pleochroic radiohalos in biotites.

MATERIALS AND METHODS

Biotites were collected from the Bear Lake Road (BLR) occurrence in Monmouth, Ontario, Canada and the Golding-Keene Quarry (GKQ) in Bancroft, Ontario, Canada, as well as from Ii Mori (IM), Japan.

The Canadian samples were all hand-sized pieces of biotite supplied by a mineralogist. At the Bear Lake Road (BLR) occurrence, Monmouth (Tory Hill), on Highway #121 approximately 24 miles west by road of Bancroft, the biotite comes from a calcite vein/dyke and is associated with calcite, apatite, amphibole and titanite. Details of the rock hosting the vein/dyke were not supplied, but 6 miles to the northeast in the Wilberforce area are calcite vein/dykes that intrude syenitic rocks (syenitized biotite-gneisses and amphibolites).[9] The biotite from the Golding-Keene Quarry (GKQ) at Bancroft comes from what is described as a nepheline pegmatite, which as well as biotite consists of nepheline, sodalite, cancrinite, hackmanite and albite. Again, details of the rock hosting the nepheline pegmatite were not supplied, but mafic and syenitic rocks, particularly nepheline syenite, intrude metasediments in the area,[10] so it is possible the pegmatite is associated with the nepheline syenite. The biotite sample from Ii Mori (IM), Japan, was supplied as a small piece by Dr Robert Gentry, who refers to Iimori and Yoshimura[11] as the source of this biotite.[12] Wise[13] found that this biotite actually comes from the Ishigure District of Japan, but unfortunately no other geological details have so far been forthcoming.

The biotite samples from each site were cleaved under a stereo microscope into 1" x 1" (approximately 2.5 cm x 2.5 cm) sections, which were then examined under a Carl Zeiss JENA compound microscope for the presence of radiohalos. Sections containing radiohalos were further cut into sizes of 1cm x 1cm for placement into crucibles and numbered. Low power (1X and 6.3X objectives) and mid-range power (12.5X and 25X objectives) photomicrographs were taken before and after heating events, and each section was hand drawn with the approximate locations of radiohalos and large physical landmarks in relation to section borders, along with an outline of the area which was photographed.

Two experimental studies were designed, one to test the effect of increasingly higher temperatures over a short interval, and the other to test the effect of a constant moderate temperature over a designated period of time. For the high temperature study, an American Scientific Products FP-21 digital muffle furnace was employed, while a Perkin Elmer Aerograph 200 furnace was used for the long duration study. Samples used in the high temperature study were placed into Vycor and platinum 50 ml crucibles, while 50 ml Pyrex flasks were employed in the low temperature study.

EXPERIMENT 1

Samples IM-1 through 1M-6, GKQ-1 through GKQ-4, and BLR-1 through BLR-5 were prepared for the high temperature study as described above. Table 1 lists the samples used, the source of each sample, the amount of time each sample was subjected to thermal stress and the range of temperatures that each sample was exposed to.

Group A

For group A (samples IM-1 to IM-6), the oven was preheated to 250°C and all samples were introduced. After one hour, two samples were removed and the oven temperature was increased by 100°C. Every hour this procedure was repeated until all samples had been removed for microscopic examination, the last sample (IM-6) being removed after the oven reached a maximum temperature of 550°C during the fourth hour.

Group B

For Group B (samples GKQ-1 to GKQ-4 and BLR-1 to BLR-5), the oven was preheated to 550°C and all sam-

Sample		Total Exposure Time	Temperature (°C)	
Number	Source Locality		Initial	Final
IM-1	Ii Mori, Japan	1 hour	250	250
IM-2	Ii Mori, Japan	1 hour	250	250
IM-3	Ii Mori, Japan	2 hours	250	350
IM-4	Ii Mori, Japan	2 hours	250	350
IM-5	Ii Mori, Japan	3 hours	250	450
IM-6	Ii Mori, Japan	4 hours	250	550
GKQ-1	Bancroft, Canada	1 hour	550	550
GKQ-2	Bancroft, Canada	2 hours	550	600
GKQ-3	Bancroft, Canada	2 hours	550	600
GKQ-4	Bancroft, Canada	3 hours	550	650
BLR-1	Monmouth, Canada	4 hours	550	700
BLR-2	Monmouth, Canada	2 hours	550	600
BLR-3	Monmouth, Canada	1 hour	550	550
BLR-4	Monmouth, Canada	3 hours	550	650
BLR-5	Monmouth, Canada	3 hours	550	650

Table 1. *Summary listing of the samples used in Experiment 1 and the experimental conditions.*

Sample		Total Exposure Time	Temperature (°C)
Number	Source Locality		
IM-7	Ii Mori, Japan	5 hours	300
IM-8	Ii Mori, Japan	5 hours	300
IM-9	Ii Mori, Japan	5 hours	300
BLR-6	Bancroft, Canada	5 hours	300
BLR-7	Bancroft, Canada	5 hours	300
BLR-8	Bancroft, Canada	5 hours	300

Table 2. *Summary listing of the samples used in Experiment 2 and the experimental conditions.*

ples were introduced. After one hour, two samples were removed and the oven temperature was increased by 50°C. Every hour this procedure was repeated until all samples had been removed for microscopic examination, the last sample (BLR-1) being removed after the oven reached a maximum temperature of 700°C during the fourth hour.

EXPERIMENT 2

Samples IM-7 through IM-9 and BLR-6 through BLR-8 were prepared for the long duration study as described above. Table 2 lists the samples used, the source of each sample and the amount of time each sample was subjected to thermal stress. The oven was preheated to 300°C and all samples were introduced simultaneously. The oven was maintained at 300°C, and after a five hour period all the samples were removed for microscopic examination.

RESULTS

Gross Damage

In experiment 1, at the lower temperatures, the IM samples were greatly affected by thermal stress. Although landmarks were intact and there was no gross change in the mineral at 250°C, the mica began to darken significantly and flake away at 350°C. At 550°C, the IM samples had darkened in some sections to the point where they were no longer transparent under 12.5X objective magnification. Landmarks within the samples underwent significant changes, either by full fracturing, flaking and splitting within the biotite, or by complete darkening (see Figures 2 and 3).

At higher temperatures, the GKQ samples reacted much like the IM samples had previously. Flaking and darkening were immediately evident after only one hour of exposure to 550°C, and simply became worse as further thermal stress was applied. One item of note is that the GKQ samples, much like the IM samples, underwent a color change from green to brown at the highest amount of exposure. Further study is needed to characterize colour changes over time at high temperatures within the biotites from different sample localities. The BLR samples, on the other hand, appeared to be more unyielding on a structural level to heat exposure than the GKQ and IM samples. Little darkening of the biotite was evidenced even after four hours of exposure from 550° to 700°C. However, significant erasure of large landmarks was evident at 600°C (see Figures 4 and 5).

In experiment 2 the BLR samples showed minor colouration differences and some landmark erasure, while the IM sections manifested major changes in landmarks, colouration and structure, although little or no flaking took place.

Microscopic Radiohalo Observations

Experiment 1

Radiation stains and halo rings began to degrade and disappear after as little as two hours at 350°C. Figures 6 and 7 show pre- and post-heat photomicrographs of sample IM-4 and its Po-218 halos at 125X magnification. The post-heat photomicrograph demonstrates the absence of the outer Po-218 ring. Po-210 halos seen in sample IM-5 (see Figures 8 and 9) are clearly not there after three hours of exposure to heating up to 450°C. It is at these temperatures that landmarks and microcracks disappear, or are altered so as to make them unrecognizable. For example, the IM-6 pre- and post-heat photomicrographs (Figures 10 and 11) show not only that the halos are annealed, but the mica has become completely altered in colour, texture and orientation. GKQ-1, which spent only one hour at 550°C, exhibited complete halo erasure (see Figures 12 and 13). Three Po-210 halos found in BLR-1 were reduced to simply the radiocenters after the four hours exposure at temperatures from 550-700°C (see Figures 14 and 15).

Experiment 2

The radiohalos fared no better under the lower temperature conditions of the second experiment. Not only were halos completely obliterated as before, but the mica was again altered significantly, as seen in sample IM-7. The IM-8 pre- and post-heat photomicrographs (Figures 16 and 17) show the erasure of Po-210 halos, with no significant mica alteration other than the major discolouration of a red mineral (hematite?) embedded within the mica. On the other hand, IM-9 showed some Po halo erasure with mica alteration. Curiously, the BLR-6 through BLR-8 specimens showed minimal heat damage and little halo erasure after the five hours exposure to 300°C.

DISCUSSION

The results of this limited study demonstrate that biotites from different localities certainly react differently to heat exposure. This probably reflects the different geological context and origin of each of the three biotite samples utilized in these experiments. Although some biotites appeared to be minimally affected, discolouration and cracking/delamination of the mica can and does occur with exposure to heat, even at temperatures as low as 300°C. Few if any of the biotites inspected by the authors while searching for radiohalos for this study exhibited such marked heat-related effects. It follows, therefore, that the biotites within which the radiohalos were found must not have been subjected to elevated temperatures for protracted periods since formation of the radiohalos. Our results also indicate that higher temperatures may be required to achieve halo erasure in some biotites, although in others erasure may be achieved without melting the biotite, or even healing fractures or other landmarks. In addition, we have found that short interval/lower temperature heating events can erase some, but not all, rings associated with a given radiohalo.

The implications of these experimental results may be far-reaching and significant. If the biotites within which these Po-218 radiohalos are found cannot have been subjected to elevated temperatures for protracted periods since formation of the radiohalos, then neither could the enclosing rocks. Yet if the GKQ nepheline pegmatite was hosted by nepheline syenite, for example, then the biotite supposedly crystallized at temperatures well in excess of 500°C, with the radiohalos forming around the included radiocenters immediately while the whole pegmatite, and perhaps even the host nepheline syenite, were cooling over thousands of years. However, this scenario must now be questionable, since the radiohalos would have been erased by such heat, given that in our experiment after only one hour of exposure to 550°C the GKQ biotite began flaking and changing colour, while the radiohalos were completely erased.

If, on the other hand, the radioactive Po-218 migrated in solution into the radiocenters after the biotite and its host rocks had completely cooled, where are all the microcracks along which the solution migrated? We found in this study no apparent microcracks or clefts associated with the radiohalos in the GKQ biotite crystals. Indeed, we found that the applied heat either largely erased such landmarks or the landmarks were further fractured, with flaking or splitting within the biotite grains resulting. Lest it be argued that the absence of microcracks and fractures associated with the radiohalos is because they were originally there but were subsequently healed, any heat which must surely have been involved in such a supposed healing process would, as our experiments have shown, not only have healed the microcracks and fractures, but erased the radiohalos that the microcracks and fractures were supposed to have assisted in producing. So our experiments potentially rule out the solutions-migrating-along-microcracks theory for the origin of these Po-218 radiohalos.

Similar arguments should apply to the BLR biotite results. If the BLR calcite vein/dyke is similar to that in the nearby Wilberforce area, then it probably intrudes, or is conformable to, syenitized biotite-gneisses and amphibolites, and was thus supposedly also formed by the same suggested hydrothermal 'fluxing' and recrystallization of wallrocks along bedding planes, joints and other fractures.[14] This of course would mean that the biotite crystallized at temperatures of at least 100° to 300°C, which supposedly persisted for a subsequent lengthy period during cooling of the vein/dyke and its wallrocks. However, in our experiments the BLR biotite samples appeared to only show minor colouration differences, some landmark erasure and little halo erasure after heating at 300°C for five hours, and little darkening even after heating for four hours from 550° to 700°C, although there was significant erasure of large landmarks at 600°C and complete erasure of Po-210 halos after the four hours of heating from 550° to 700°C. Thus the possibility exists, at least in this instance, that the radiohalos in this biotite may have survived the presumed ambient temperatures at the time of their formation. Nevertheless, the presumed timescale of hundreds or thousands of years for biotite and pegmatite formation still cannot accommodate radiohalo formation from Po-218, an isotope with a fleeting existence, while again no microcracks or fractures were found associated with the Po radiohalos in our BLR biotite samples that would have allowed solution transport of Po to the radiocenters. In any case, such solution transfer of Po-218 from an undesignated source over an undesignated distance to these particular biotite flakes (and no others — for example, in the biotite-gneiss wallrocks) would still need to have been remarkably (impossibly) rapid — less than 5 days according to Feather,[15] who concluded that such an hypothesis *'cannot be accorded very great credulity'*.

Figure 2. *Photomicrograph of biotite in sample IM-6 prior to heating (63X magnification).*

Figure 3. *Photomicrograph of the same biotite in sample IM-6 after heating 250–550°C over 4 hours (63X magnification). Damage to the biotite has been considerable.*

Figure 4. *Photomicrograph of biotite in sample BLR-2 prior to heating (63X magnification).*

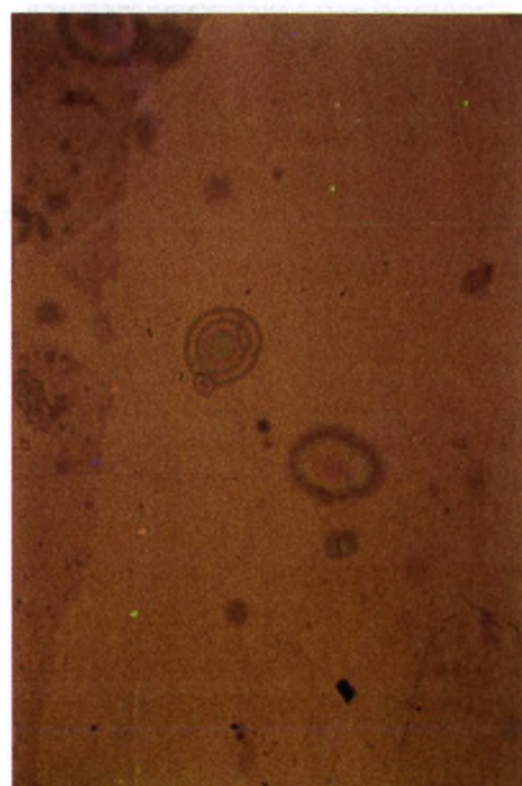

Figure 5. *Photomicrograph of the same biotite in sample BLR-2 after heating to 600°C over 2 hours (63X magnification). Large landmarks have been erased.*

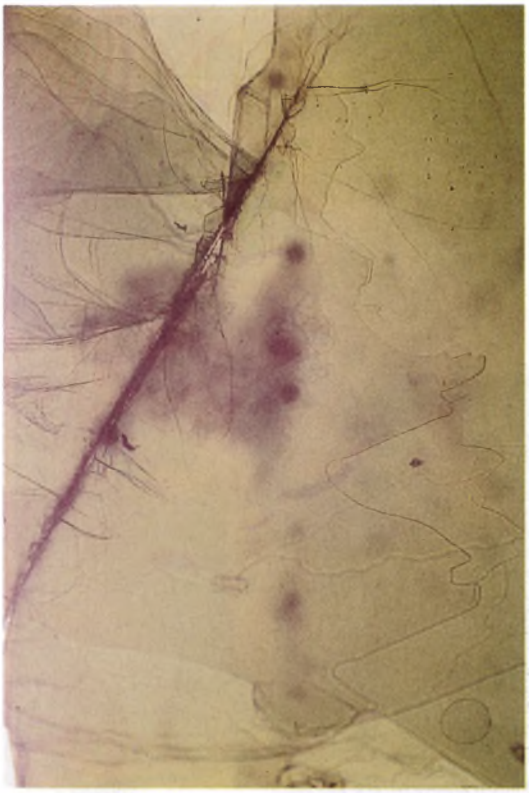

Figure 6. *Photomicrograph of biotite with Po-218 radiohalos in sample IM-4 prior to heating (125X magnification).*

Figure 7. *Photomicrograph of the same biotite and Po-218 radiohalos in sample IM-4 after heating 250–350°C over 2 hours (125X magnification). Some halo ring erasure has occurred.*

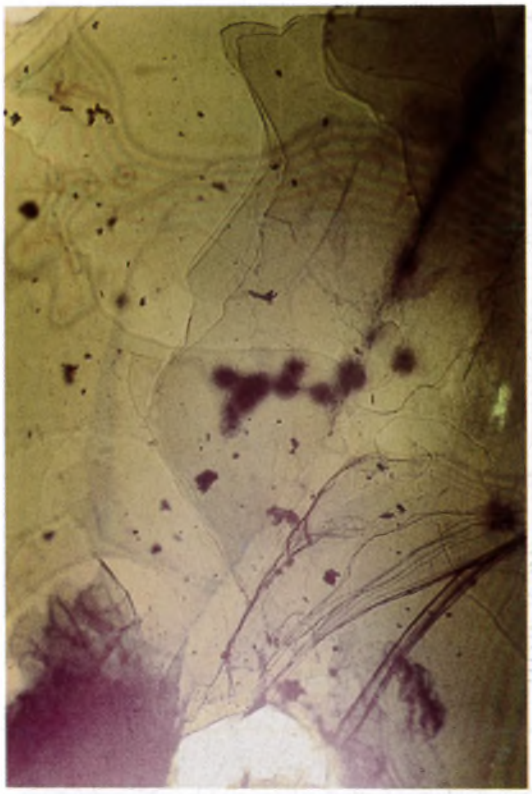

Figure 8. *Photomicrograph of biotite with Po-218 radiohalos in sample IM-5 prior to heating (125X magnification).*

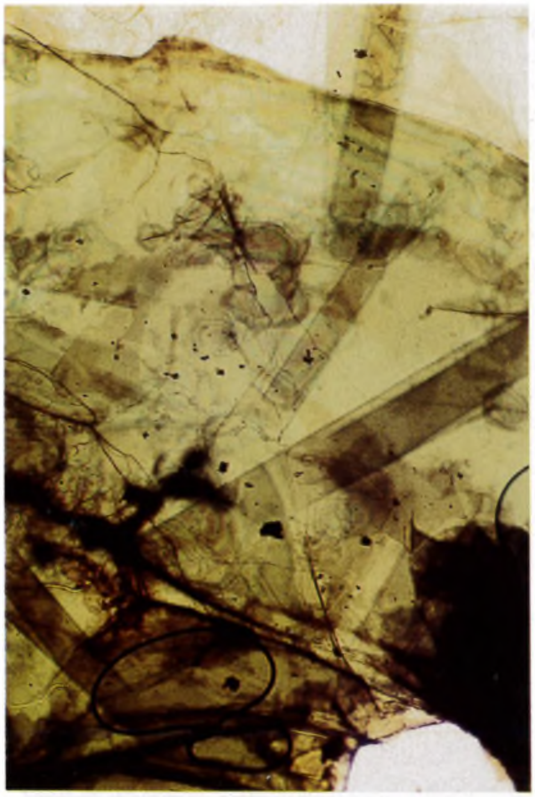

Figure 9. *Photomicrograph of the same biotite in sample IM-5 after heating 250–450°C over 3 hours (125X magnification). The Po-210 radiohalos have been erased.*

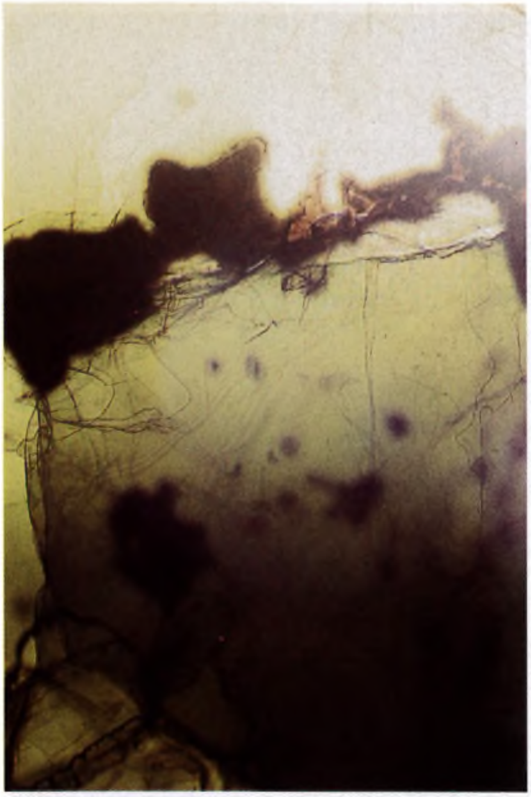

Figure 10. *Photomicrograph of biotite with Po-218 radiohalos in sample IM-6 prior to heating (125X magnification).*

Figure 11. *Photomicrograph of the same biotite and Po-218 radiohalos in sample IM-6 after heating 250–550°C over 4 hours (125X magnification). The damage and erasure are severe.*

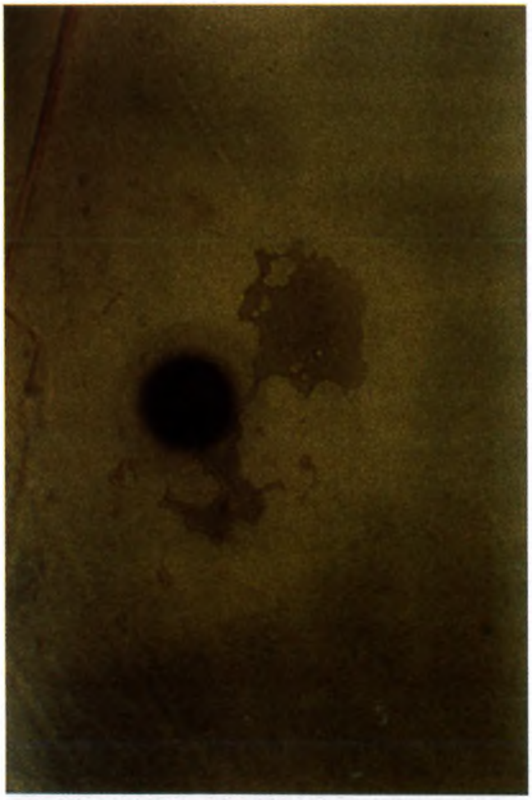

Figure 12. *Photomicrograph of biotite with Po-218 radiohalos in sample GKQ-1 prior to heating (250X magnification).*

Figure 13. *Photomicrograph of the same biotite in sample GKQ-1 after heating at 550°C for 1 hour but the Po-218 radiohalos have been erased (250X magnification).*

Figure 14. *Photomicrograph of biotite with Po-210 radiohalos in sample BLR-1 prior to heating (125X magnification).*

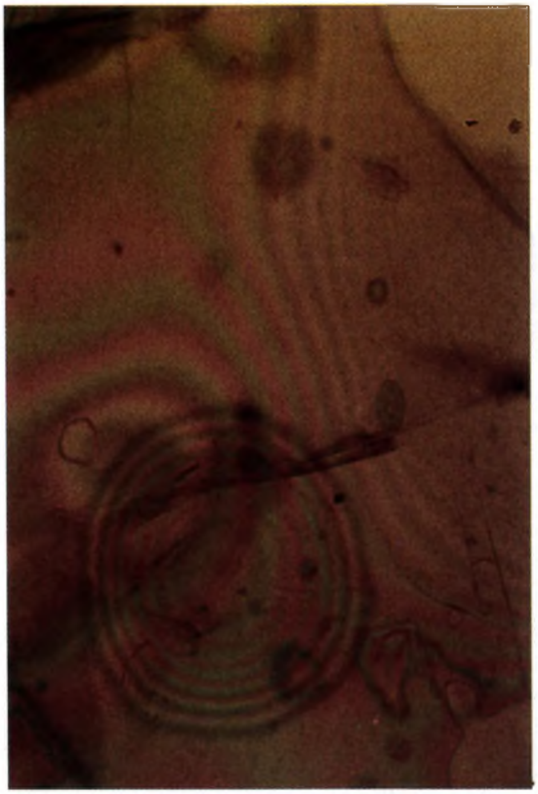

Figure 15. *Photomicrograph of the same biotite in sample BLR-1 after heating 550–700°C over 4 hours (125X magnification). The radiohalos have been erased but the radiocenters remain.*

Figure 16. *Photomicrograph of biotite with Po-210 radiohalos in sample IM-8 prior to heating (125X magnification).*

Figure 17. *Photomicrograph of the same biotite in sample IM-8 after being heated at 300°C for 5 hours but the Po-210 radiohalos have been erased (125X magnification).*

These experiments, therefore, may not 'prove beyond reasonable doubt' that the biotites and host rocks containing these Po radiohalos crystallized very rapidly, but they do put specific constraints on both the role of heating/hot processes and the timescales involved. These, in turn, raise serious questions about the validity of the conventional wisdom of igneous, metamorphic and/or hydrothermal processes of formation for these biotites and their host rocks, all of which are supposed to have involved elevated temperatures over considerable (to long) periods of time. Similarly, our observations and experiments appear to quite specifically rule out the solutions-migrating-along-microcracks theory of Po radiohalo formation subsequent to biotite and host rock crystallization. Can it be that these Po radiohalo-bearing rocks not only formed very rapidly, but under cold conditions?

There may be one other possibility. Baumgardner[16] has suggested that nuclear decay rates may not have always been constant during the earth's history. He reasons that it is difficult to imagine what mineralization process could yield tiny mineral grains (radiocenters) with high concentrations of Po inside much larger grains of other minerals in a granitic rock on a timescale short enough so that sufficient Po (or its beta precursors) remained after the rock became cool enough for radiohalos to form, unless the nuclear decay rates when the granite crystallized were much lower than at present. In other words, if granites (and pegmatites, vein/dykes, etc.) were once in molten form and the Po present had to be isolated and concentrated during the magma crystallization process, then the nuclear decay rates had to be much slower so that the Po had much more than a fleeting existence. Nevertheless, while this is an intriguing possibility, with tantalizing implications with respect to radioactive dating of rocks and minerals, our experiments still demonstrate that heat, even over a matter of a few hours, can erase these Po radiohalos, which still throws doubts on the role of heat and hot processes in the formation of these Po radiohalo-bearing biotites and their host rocks.

Further experimental work is undoubtedly required before such conclusions can be drawn with any finality. In particular, it would be ideal to test suites of biotite samples from a larger number of better characterized geological situations (for example, from granites and granite pegmatites), and to subject such samples to a wider range of temperatures (for example, 100° to 700°C) over longer periods, perhaps even over days or weeks. Such a wider study should provide clearer correlations and patterns of responses of the biotites and the Po radiohalos to temperatures and timescales, and with respect to biotite and host rock types/geological contexts.

CONCLUSIONS

The exposure of Po radiohalo-bearing biotites to heat from 250° to 700°C for up to five hours causes variable but significant changes and damage to biotites, and can erase both structural defects/landmarks and radiohalos. This evidence supports our contention that the elevated temperatures and long timescales conventionally postulated for the igneous, metamorphic and/or hydrothermal processes supposedly responsible for the formation and subsequent cooling of these biotites and their host rocks cannot account for the occurrence of these Po radiohalos. Furthermore, our observations and experiments discount the other explanation often given, that solutions migrating along microcracks subsequent to biotite formation deposited the Po isotopes in the radiocenters. Thus it is possible that the textbook-popularized hot magma scenario for the formation of some crystalline rocks may not be a true representation, and that these Po radiohalo-bearing biotites and their host rocks may have formed/crystallized very rapidly, possibly even under cold conditions. Alternately, nuclear decay rates may have possibly been much slower when crystallization occurred.

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