Radioactive halos as possible indicators for geochemical processes in magmatites

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(Received August 9, 1976)

In systematic microscopic measurements completed by fission track studies the occurrence of polonium halos and polonium bands could be confirmed in biotites of different areas. The abundance of the different types of polonium halos is given by the order $^{210}$Po $>$ $^{218}$Po $>$ $^{214}$Po. As magmatic and often also hydrothermal mechanisms are contrary to geological evidences a supergene hypothesis has been supposed for the genesis of the polonium halos. According to this mechanism polonium isotopes are accumulated out of uranium- or radium-bearing solutions which are liberated from rocks by leaching processes of percolating weathering waters. The concept of the given origin of polonium halos may be significant in discussing problems of dating, uranium prospection and the origin of secondary uranium deposits.

INTRODUCTION

Radioactive halos which are produced in minerals as concentric shells of coloration by alpha particles of radioactive inclusions have found renewed attention for the last few years (Gentry, 1973) in spite of their discovery and explanation already at the beginning of this century (Joly, 1907; Mügge, 1907; Hövermann, 1912).

This interest results from the correspondence between the radii of halos and ranges of alpha particles and from the association of halo dimensions with alpha energies of decaying radionuclides. The halos, microscopic defects in the lattice of the minerals caused by alpha particles are very stable and stored in minerals of ancient rocks over long periods. Therefore, it is possible to find alpha emitters of the decay series of $^{238}$U or $^{232}$Th in microscopic inclusions from the Precambrian up to the Tertiary, which might be of interest in chronological estimations (Longinelli, 1960; Przibram, 1953; Meier, 1966; Hirschmann, 1967) or in an examination of the constancy in time of the radioactive decay (Gentry, 1973; Spector, 1972). Moreover, it is possible to get - from anomalous halos - hints at extinct radionuclides or at the existence of radionuclides which are unknown up to now. For instance, the discovery of extinct radionuclides of the $^{237}$Np series could give an insight into the solidification time of the earth (Ramdohr, 1957), or the detection of halos of anomalous radius which cannot be attributed to known alpha emitters would offer interesting aspects in the world-wide search for natural superheavy elements (Ader, 1972; Gentry, 1966a, b, 1970, 1971).

In addition to halos which cannot be associated with known naturally occurring alpha emitters the existence of a special group of radiohalos, the so-called polonium halos, have been discussed since the 30's (Henderson and Sparks, 1939; Henderson, 1939; Gentry, 1968, 1974; Gentry et al., 1973, 1974). In spite of the possibility of coordinating ring sizes with well-known alpha emitting polonium isotopes there are serious difficulties in a convincing explanation of these halos. This results from the lack of rings belonging to any parent nuclide of the uranium series which should be expected because of the short half-lives of polonium isotopes. Therefore, the origin of polonium halos has been controversial. And even in a recent work, the existence of polonium halos has been generally questioned (Moazed et al., 1973).

Because of this contrariety a systematic study has been started in order to elucidate problems concerning the existence and the origin of polonium halos.
EXPERIMENTAL

In spite of the occurrence of halos in various minerals (RAMDOHR, 1957; STARK, 1936) in this study we have examined mainly biotite samples. We selected biotite because this mineral can be considered as a good halo detector with a high transparency in thin sections and good preparation conditions. Moreover, the existence of polonium halos has been reported mostly in mica (biotite) samples from different localities. In this work biotite samples mainly from Kragerø, Froland and Moss (Norway), and Faraday Township (Ontario, Canada) have been studied.

For obtaining real radii of halos which correspond to alpha particle ranges it must be noted that the radiation-damaged area is shaped as a sphere with the alpha emitters in its center. Therefore, only the radii of halo rings obtained from a plane section through the center of this sphere can be used for the determination of the range of the alpha particles. From plane sections which contain only the parts of the halo above or below this diametral section may result misleading radii and alpha ranges, respectively, (see Fig. 1).

![Fig. 1. Scheme for the determination of the “true diameter” of radiohalos.](image)

That means, an exact determination of radiohalos often needs measurements of halo radii in different sections of spheric halos. In this manner the diametral section is fixed as a maximum in a series of rings with increasing and decreasing sizes. With biotite which is characterized by ideal cleavage properties this can often be done in a relatively simple manner by successive lifting of mica layers with a transparent adhesive tape; see also (GENTRY, 1973).

In the presence of microscopically visible inclusions of, e.g., accessory minerals such as zircon, monazite, xenotime and others (GENTRY, 1973; SNETSINGER, 1967; SCHWANDER and WENK, 1965) the ring sizes can be considered as well-defined if these inclusions are situated in the center of a halo. However, it should not be overlooked that longshaped or large (0.2µm) inclusions with adsorbed or built-in radio-nuclides can produce elliptical halos (GENTRY, 1971) or halos with anomalous sizes.

As mentioned above the ranges of alpha particles emitted from an invisible cluster of at least $10^8$ - $10^9$ radioisotopes (GENTRY, 1973) or from a radioactive inclusion are measured with the real radii of halos. Moreover, the energies of those alpha emitters can be derived from range-energy curves which are given, e.g., for mica as a host mineral (GENTRY, 1967); see Fig. 2.

![Fig. 2. Range-alpha energy curve for mica (according to GENTRY (1967)).](image)

For scrutinizing biotite samples upon thin slicing and for an exact ring analysis a Leitz Nuclear-Track Microscope and a Leitz Orthoplan Microscope with magnifications between 100 and 625 have been used. The measurements of well defined halos were made with a micrometer ocular with an error of about ± 1µm.

The results of these optical microscopic studies have been completed by gammaspectroscopical determinations of U, Th and K concentrations and by determinations of the distribution of uranium in biotite samples with the help of fission-track techniques (HF etching and microscopical identification). Moreover, electron-induced X-ray fluorescence analytical measurements (MEIER et al., 1970) of radiocenters of polonium halos as described in GENTRY (1974) and GENTRY et al. (1974) have been started in a similar manner; the results, however, will be reported in another paper.

RESULTS

By systematic optical measurements of specimens consisting mostly of biotite some halos with rings attributable to the alpha decay of
$^{146}$Sm ($E_a = 2.2 \text{MeV}$) and the members of the $^{238}$U and/or $^{232}$Th series, have been observed. The greatest portion of halos, however, could be clearly identified as polonium halos. In this context it should be noted that polonium halos are defined as halos which seem to result from the decay of polonium isotopes of the $^{238}$U series without any visible connection to other alpha emitting nuclides of the $^{238}$U series. Since three polonium isotopes, i.e. $^{210}$Po and $^{214}$Po and $^{218}$Po, are members of the $^{238}$U series, the alpha decay of halos can principally start from $^{218}$Po, $^{214}$Po or $^{210}$Po. Therefore, in accordance with ranges and energies, resp., of alpha particles emitted from these isotopes (see Table 1) which are decaying in the series $^{218}$Po $\rightarrow$ $^{214}$Po $\rightarrow$ $^{210}$Po, several structures of polonium halos can be expected.

I.e., the different types of polonium halos are:

- $^{218}$Po halos which are characterized by a three-ring structure due to the decay of three isotopes;
- $^{214}$Po halos which are characterized by a two-ring structure due to the decay of two isotopes;
- $^{210}$Po halos which consist only of a disc.

In Fig. 3 these distinct types of polonium halos are schematically shown; see also (GENTRY, 1973; HENDERSON and SPARKS, 1939; MOAZED et al., 1973).

Moreover, some observations have been made which may be useful for the explanation of the origin of these radiohalos:

1. In the class of polonium halos the most common ones belong to the $^{210}$Po type; $^{218}$Po halos have been found in a less portion whereas $^{214}$Po halos are very seldom. From Table 2 which has been taken from measurements of biotite (Kragerø, Norway) this distribution of different types of polonium radiohalos can be derived. This result agrees also with Henderson’s observation (HENDERSON, 1939) that $^{210}$Po halos (type A according to HENDERSON) are commoner than $^{218}$Po halos (type C).

2. By a fission-track analysis of the centers of polonium halos no tracks resulting from spontaneous fission of $^{238}$U could be observed in most halos; see Figs. 6 and 7. Therefore,
Table 2. Distribution of radiohalo types found in biotite (Kragerø, Norway)

<table>
<thead>
<tr>
<th>Radiohalo type</th>
<th>Number of halos observed</th>
</tr>
</thead>
<tbody>
<tr>
<td>Po-210</td>
<td>1,000</td>
</tr>
<tr>
<td>Po-218</td>
<td>90</td>
</tr>
<tr>
<td>Po-214</td>
<td>1</td>
</tr>
</tbody>
</table>

it can be stated that no or only small amounts of uranium are present in polonium halos, a result, which agrees with the lack of uranium ring-structures in these halos; see also (GENTRY, 1968). In this context one should remember that fission track studies allow a determination of uranium contents down to the ppm range (WAGNER, 1973). Moreover, biotites are suitable for these studies (WELIN et al., 1972) and an error by track fading can be excluded in most cases because fission tracks have been observed in other areas within the same specimens, even in the nearest neighbourhood.

3. The results of petrologic studies which have shown that the nuclei of uranium or thorium halos are usually formed in biotite by small accessory minerals such as apatite, zircon, monazite or xenotime (SNETSINGER, 1967; OSBORNE, 1947) can not be generally applied to the centers of polonium halos: There are a lot of polonium halos without any microscopic visible center. Furthermore, polonium halos are often found located at defects of mica, i.e. at cracks, veins, microscopic structural distortions or conduits. In fluorite samples Po halos could be found - analogous to SCHILLING (1926) - only along cracks and never in undisturbed specimens; but see GENTRY (1973).

These observations point to an important difference between U and Th halos, on the one hand, and Po halos on the other hand: Whereas the genesis of U and Th halos is connected with an inclusion of uranium or thorium nuclides into the lattice of small accessory minerals during their crystallization from the magma and before the later crystallization of biotite, polonium radiohalos are not formed by an entry of polonium isotopes into the lattice of accessories during the magmatic crystallization. The observation of an accumulation of polonium halos at distorted areas and cracks of mica suggests that polonium isotopes must be deposited at defects of mica at a later stage.

That is to say, it can be supposed that those polonium halos which are connected with small accessories might result from an adsorption of polonium isotopes on inclusions. Moreover, the tendency for the occurrence of polonium halos at fine cracks and conduits can result in an accumulation of clusters of polonium halos which may lead to coloration bands with radial extensions given by the diameter of polonium halos. As an example, in Fig. 8 corresponding “polonium bands” are given. In this context, the “spectacle” coloration pattern found by GENTRY et al. (1974) in a biotite sample should also be mentioned.

4. A comparison between spontaneous fission-tracks of $^{238}$U and polonium halos showed in biotite samples no correlation between the distribution of tracks and polonium halos (see Fig. 6). However, many samples showed that a high density of polonium halos is often connected with a high density of uranium fission tracks. In accordance with this observation measurements of the radioactivity of mica matrix gave obvious evidence for the fact that samples rich in polonium halos belong to min-

Fig. 6. Example for demonstrating the fact that Po-halos are not connected with fission-tracks, i.e., with $^{238}$U (Note: etching conditions HF (1 : 2), 10min 20°C; magnification 290 X; see also Fig. 8 before etching).

Fig. 7. Fission tracks from spontaneous fission of $^{238}$U obtained in a distance of 0.3mm from the $^{210}$Po-halos given in Fig. 6.
erals with high uranium concentrations; e.g., the uranium concentration of the biotite of Kragerø mentioned above is in the order of 24 ppm. The uranium content of the mica samples studied, however, can not be ascribed to accessory minerals with high uranium concentrations as indicated by fission track measurements: A large portion of fission tracks whose density is approximately proportional to the uranium content are concentrated upon dislocations and microscopic cracks.

That is, the origin of polonium halos in biotites and other minerals must be related in any way above all to the presence of uranium which is not associated with accessory minerals but concentrated by adsorption, isomorphous substitution etc. in rock-forming minerals as discussed above for biotites (LEONOVA and TAUSON, 1958).

5. By ion microprobe mass spectroscopic studies of polonium halo inclusions, GENTRY (1974), GENTRY et al. (1973), GENTRY et al. (1974) and MOAZED et al. (1973) yielded results exhibiting anomalously high $^{206}\text{Pb}/^{207}\text{Pb}$ isotope ratios which are not consistent with the Pb isotopic ratios in U and/or Th halos. Since these Pb isotopic ratios can be considered as a consequence of the decay of polonium isotopes, such as $^{218}\text{Po}$, to $^{206}\text{Pb}$ an obvious evidence for the different kind of Po and U and/or Th halos is given. We could not find Pb in the centers of polonium halos by electron-probe X-ray analysis up to now. However, this negative result may perhaps be explained by difficulties in the preparation process for obtaining halo centers at the surface of cleavage layers; see also GENTRY et al. (1974).

**Discussion**

Every hypothesis concerning the origin of polonium halos is confronted with the fact that the halo-forming polonium isotopes have short half-lives of the order of $10^{-4}$ seconds to days. Moreover, any mechanism must be consistent with geological facts.

1. **Magmatic and hydrothermal mechanisms of Po halo formation**

   **A. Magmatic hypothesis**

   According to the magmatic hypothesis the formation of Po halos should be the result of an inclusion of polonium isotopes in rock-forming or accessory minerals during the process of magmatic crystallization. This mechanism would only be possible in quickly crystallizing systems. Because of the slow magmatic cooling rates of polonium-halo-bearing rocks such as granites, however, this hypothesis is untenable.

   The contrariety to the geological evidence could be overcome with long half-life isomers of polonium of $\beta$-decaying precursors as discussed by GENTRY et al. (1973). However, the ion-microprobe search for the isomers which should still exist in measurable quantities brought no positive results up to now (GENTRY, 1974; GENTRY et al. 1973, GENTRY et al., 1974) so that this hypothesis has been abandoned (GENTRY, 1974).

   **B. Hydrothermal hypothesis**

   In this hypothesis postulated by HENDERSON (1939) polonium halos are supposed to be of hydrothermal origin. Therefore, the formation of halos is discussed as an accumulation process of polonium isotopes out of uranium-bearing hydrothermal solutions at different deposition centers of previously crystallized biotite. As hydrothermal solutions coming from a magma reservoir may be transported through biotite by a linear or laminar flow, polonium halos should be localized above all along narrow conduits and veins or in certain leaves near these clefts.

   In contrariety to this hypotheses GENTRY (1968) cites measurements of the alpha-recoil density showing no excess alpha activity near polonium halos which should be expected in the presence of alpha-decay precursors in hydrothermal fluids. Moreover, polonium halos cannot be ascribed only to biotite samples which have been in contact with hydrothermal solutions during their geological history.

   **2. Po halo genesis as a consequence of weathering processes**

   **A. Principle of the supergene hypothesis**

   The accumulation hypothesis of HENDERSON
(1939) can be considered as a valuable basis for an explanation of the genesis of polonium halos. However, the assumption of a supply of radio-nuclides into rock-forming minerals by late magmatic or hydrothermal fluids restricts this kind of halo genesis to special localities. Therefore, for an explanation of the genesis of polonium halos besides hydrothermal processes the influence of weathering and supgene processes, resp., should also be taken into account. According to this supgene model polonium isotopes are accumulated out of uranium bearing or radium bearing solutions which obtained their uranium or radium from rocks, such as granites, through leaching processes by percolating natural waters.

B. Experimental facts for the given hypothesis
The following points seem to favour the given hypothesis.

a. Equilibrium condition
The uranium of igneous rocks can be regarded as being in equilibrium with its daughter products before weathering processes occur by infiltration of aerated water. Therefore, at the start of weathering processes by the dissolution of microscopic and submicroscopic uranium-bearing inclusions (LEONOVA and TAUSON, 1958; BARRANOV et al., 1962) this equilibrium must not noticeably be disturbed in slow moving solutions.

However, because of the difference in the mobilities of uranium (as UO$_2^{++}$) and its daughter products under supgene conditions a disequilibrium in the decay chain of uranium may occur by a separation of the longest-lived uranium isotopes (half-lives of $^{238}$U : 4.5 x 10$^9$y; $^{234}$U : 2.47 x 10$^5$y) from their daughter products. This disequilibrium may result, for instance, from differences in the solubilities of $^{226}$Ra$^{++}$ and UO$_2^{++}$, from the tendency of thorium ($^{230}$Th$^{4+}$) to hydrolyze in insoluble hydrous oxides or to be quickly adsorbed on special inclusions, or from a migration of gaseous $^{222}$Rn away from its parent (DAVY, 1974; KIRBY, 1974). A series of studies showed that, because of this different migration capacity of the isotopes of uranium and its decay products, the isotopes of radium may often occur in special water-filled capillaries such as dislocations of the crystal lattice, cracks, fissures and holes (STARIK and POLEVAYA, 1957; SICHETPOTEVA, 1957).

It is important that as a consequence of this elemental separation, produced by leaching processes, the original radioactive equilibrium of the uranium series which was first established in the crystal lattice, may be replaced in the water-filled veins, capillaries etc. by new part-equilibriums starting from daughter isotopes. For instance, it seems possible that in radium-bearing solutions separated largely from uranium, an equilibrium is established in relatively short time between radium and its subsequent daughter products, among others with the polonium isotopes (Faul, 1954).

As in equilibrium for each decaying parent atom, on the average also one of each intermediate daughter atom decays, it can be written approximately (MEIER, 1966)

\[
\lambda_i N_i = \lambda_{218} N_{218} = \lambda_{214} N_{214} = \lambda_{210} N_{210} \quad (1)
\]

or

\[
\frac{0.693}{T_1} N_i = \frac{0.693}{T_{218}} N_{218} = \frac{0.693}{T_{214}} N_{214} = \frac{0.693}{T_{210}} N_{210} \quad (2)
\]

with $\lambda_i$ = decay constants, $T_i$ = half-lives, $N_i$ = number of atoms. According to eq. 2 the number of atoms of each intermediate daughter products, i.e. of $^{218}$Po, $^{214}$Po and $^{210}$Po, will be in direct proportion to its half-life, if the corresponding part of the uranium series is in equilibrium. Therefore, the number of polonium atoms in the unit volume of the radium- or uranium-bearing solutions - and in addition also in radon containing solutions (and gases) - can be estimated and, as a consequence, the graduation of the quantities of polonium isotopes in such solutions can approximately be given by:

\[
^{210}$Po $\approx$ $^{218}$Po $\approx$ $^{214}$Po \quad (3a)
\]

because

\[
T_{210} (140 \text{ d}) \approx T_{218} (3 \text{ min}) \approx T_{214} (10^{-4} \text{ sec}). \quad (3b)
\]

If the formation of polonium halos is supposed to be analogous to the hydrothermal hypotheses, the result of adsorption or precipitation of polonium atoms from these slowly flowing solutions at special points such as impurities, submicroscopic inclusions etc., and the tendency for the genesis of different types of polonium halos should be given by the same order. This graduation should also approximately be maintained by a disturbance of the flowing solutions
resulting from precipitation of Po-isotopes and Pb- and Bi-isotopes of the uranium series at different points in the capillaries or veins. However, it is possible that because of a different rate of reestablishing of $^{210}$Po on the one hand, and of $^{218}$Po and $^{214}$Po on the other hand, the number of the short-lived Po isotopes might be intensified in relation to $^{210}$Po; see also HENDRISON (1939).

Moreover, by preferable precipitation of $^{214}$Pb and/or $^{214}$Bi separation between $^{218}$Po and $^{214}$Po is possible: Because of its short half-life $^{214}$Po resulting from decay of $^{214}$Bi cannot be released from its precipitated parent isotopes. Therefore, on the Bi/Pb site $^{214}$Po halos may be formed, whereas dissolved $^{218}$Po ions rapidly generated from its short-lived parents are able to form the observed $^{218}$Po radiocenters at another point (see Fig. 9).

In spite of these disturbances it can be assumed that the order of magnitude of the tendency to form polonium halos of the different types is generally given in a crude manner by eq. 3a. Therefore, the observed distribution of the different types of polonium halos (see Table 1) in biotites which is in agreement with this graduation seems to confirm this hypothesis.

b. Leaching processes and migration ability of U, Ra

The mobility of uranium in rocks and minerals which are in contact with aerated waters is considerably high. Therefore, considerable effects of displacement or leaching of uranium are often observed in uranium-bearing rocks even in a cold wet climate (DAVY, 1974; KIRBY, 1974; TITAYEVA et al., 1973; LARSEN et al., 1955; STERN and STIEFF, 1959; STARIK, 1956; BROWN and SILVER, 1955).

In accessory minerals such as zircon or monazite uranium remains incorporated in its original structure also during effective erosion processes (KIRBY, 1974; HURLEY and FAIRBAIRN, 1957). Therefore, it is important for the mobilization of a considerable amount of uranium that biotites, which are preferentially associated with polonium halos (e.g. 20,000 - 30,000 $^{210}$Po and $^{218}$Po halos per cubic centimeter (GENTRY, 1968)), contain uranium in an amount additional to its presence as an isomorphous constituent of accessories. Autoradiographic studies showed that the main mass of uranium might be in biotite in a nonisomorphous, molecularly disseminated state and in microscopic and submicroscopic isolation of uranium minerals (LEONOVA and TAUSON, 1958; BARANOV et al., 1962; WILL, GALLIS, 1970). Therefore, it can be assumed that uranium, present in biotite in this state, may be leachable and responsible for secondary changes of the uranium contents in mica caused by weathering.

The results of autoradiographic studies (LEONOVA and TAUSON, 1958; BARANOV et al., 1962; BARTHEL and MEHNERT, 1970), fission track distribution measurements (WELIN et al., 1972; LAKATOS and MILLER, 1973) and leaching experiments (LEONOVA and TAUSON, 1958; BARANOV et al., 1962) gave clear evidence for the direct
displacements of uranium in biotite. In this context the observation of clusters of fission tracks and coloured pure uranium bands (see, e.g., Fig. 10) resulting from an accumulation of $^{238}\text{U}$ along fracture lines etc. of biotite and the occurrence of polonium halos in the neighbourhood and at the end of these U-bands, respectively, should be mentioned. This observation indicates the influence of secondary bleaching processes which are connected, on the one hand, with a migration of uranium, and, on the other hand, with a separation of uranium from polonium. Moreover, the occurrence of halos of the sizes of pure uranium rings corresponding to uranium which is not in equilibrium with its daughter products points to a migration period which may be in the example given in Fig. 10 not before one million years.

![Fig. 10. Example of uranium bands with very weak hints for Po (Biotite, Kragerø, Norway), magnification 290 X.](image)

It is remarkable that biotite can be considered to be an important uranium concentrating mineral in granites. Up to 15 to 20% of the uranium content of rocks may be sometimes found in biotite (LEONOWA and TAUSON, 1958). Therefore, by leaching processes large amounts of uranium may be mobilized in granites (e.g., in an area of 400 km$^2$ on the order of a million tons (BARBIER, 1974) resulting in uranium-bearing solutions which are flowing through veins, capillaries and cracks of rock-forming minerals as mentioned above.

Contrary to uranium which is very mobile and carried away in rock-forming minerals as $\text{UO}_2^{++}$ the secondary formed radium ($^{226}\text{Ra}^{++}$) may be concentrated near its parent source by the precipitation of its direct parent, $^{230}\text{Th}$, or by its low solubility in carbonate- and sulfate-rich waters (KIRBY, 1974; HARRISON and TAYLOR, 1966). However, systematic experimental research has also shown the migration ability of radium under natural conditions resulting in radium-bearing waters in capillaries, cracks, veins etc. by leaching from rocks (SACHEPOT'EVA, 1957; STERN and STIEFF, 1959; STARIK, 1956; HARRISON and TAYLOR, 1966). Since after a relatively short period of time radium coexists in equilibrium with its polonium daughter products, the conditions for the formation of polonium halos may be fulfilled similarly to uranium-bearing solutions.

c. Polonium accumulation

As shown above, geochemical studies give evidence for the tendency of uranium to pass from rock-forming minerals to natural waters. In addition, the migration ability of uranium and radium in natural solutions is proved. Since the geochemical mobility of the elements strongly depends on the environmental and hydrodynamic conditions, uranium and radium can be transported different distances before precipitating or co-precipitating (DAVY, 1974; KIRBY, 1974; STARIK, 1956; HARRISON and TAYLOR, 1966; DALL'AGLIO, 1971). In this context the tendency of polonium isotopes to be strongly adsorbed on impurities or to form colloids in slightly acid or neutral solutions, eventually by hydrolyzing to hydroxides or by an interaction with an impurity in the solution (HAISSINGSY and TUCK, 1964; KEPAK, 1974) should be mentioned.

According to this behaviour it is natural to expect a high tendency of polonium isotopes to accumulate and to concentrate locally by slowly flowing uranium- or radium-bearing solutions at defects or small inclusions of impurities in veins, fissions or cracks or disturbed layers of the rock-forming minerals during a long period of time. Since the formation of radium colloids of other elements seems to be connected to special conditions, for instance, to the exchange of radium with barium in barite during its migration (STERN and STIEFF, 1959), the preferential generation of polonium halos can be understood.

3. Consequences

In conclusion it should be emphasized that the hypothesis given for the generation of polonium halos is in agreement with geochemical and experimental facts. However, a series of conditions concerning the form of the occurrence of the radionuclides and the migrational ability have to be fulfilled. Therefore, it seems possible to use polonium halos as an indicator
for secondary processes concerning microscopic or macroscopic uranium migration in rock-forming minerals such as biotite.

This concept may be significant in discussing problems of dating, uranium prospection and the origin of secondary deposits:

1. It is known that the fission track ages of biotite and other minerals may be discordant with the real age of the mineral determined by the K-Ar or Rb-Sr methods (Meier, 1966; Wagner, 1973; Welin et al., 1972; Osborne, 1947; Lakatos and Miller, 1973; Bigazzi et al., 1971; Miller and Jäger, 1968; Miller, 1968; Gupta et al., 1971). For instance, younger fission track ages are caused by thermal annealing of fission tracks (Haack and Potts, 1972) in accordance with Eq. 4 (by decrease of $\rho_s$), whereas an uranium depletion by leaching percolating waters may bring about a fission track age older than the real age of the mineral (by decrease of $\rho_i$):

$$t = 6.57 \times 10^9 \log \left(1 + \frac{\rho_s \psi}{1.08 \times 10^{-7} \rho_i}\right)$$

($t =$ fission track age in years, $\rho_s =$ density of fossil $^{238}$U tracks; $\rho_i =$ density of induced $^{235}$U tracks; $\psi =$ integrated thermal neutron flux).

Since several factors may cause errors of fission track ages, the observation of polonium halos seems to be useful in the interpretation of fission track ages of mica samples: It is evident that the occurrence of polonium halos indicates the possibility of discordant fission track ages as a result of leaching effects by percolating waters which may be older (or younger) than the real ages corresponding to a loss (or addition) of uranium in different areas of rock-forming minerals. Perhaps, by a comparison with isotopic data an insight into the efficiency of leaching effects seems possible. Moreover, in such mica large cleavage layers which are characterized by an uniform uranium distribution (Gupta et al., 1971) and, as a consequence, which are not disturbed by halos may be eventually suitable for dating.

2. As only a small fraction of uranium reserves (about 10%) is represented by primary hydrothermal deposits the task of prospecting will be done in future above all by detecting secondary deposits (Hain-Wenniemeier, 1972, Fuchas and Siegers, 1970). Therefore, uranium occurrences which may be formed through hydrogeochemical weathering of crystalline rocks, predominantly granites, need attention (Barbier, 1974; Dall’Aglì et al., 1974; Maucher, 1962). If uranium deposits are considered to be the result of mobilization of uranium, above all from granites, followed by precipitation and accumulation processes the secondary deposits may be quite distant from the uranium-bearing motherrock. Since the presence of polonium halos in rock-forming minerals, such as biotite, on a sample-scale can be discussed as an evidence for efficient leaching processes which may concern large areas, therefore, conclusions about the possibility of finding uranium-bearing deposits in the vicinity should be taken into account.

3. As discussed above the origin of polonium halos in biotites from different areas cannot generally be attributed to hydrothermal processes. However, the supergene hypothesis which is connected with an efficient displacement and leaching of uranium by percolating waters seems to be able to explain the genesis of this type of radiohalos. It is interesting to note that this mechanism is in agreement with the hypothesis of the formation of secondary uranium deposits by continental weathering as given by Barbier (1974) and Dall’Aglì et al. (1974). Therefore, the observation of polonium halos in biotites and other rock-forming minerals should be included in the discussion concerning the genesis of uranium deposits by hydrothermal processes on the one hand, and supergene processes on the other hand.

The given concept and its consequences will be tested in further studies.

Acknowledgments—The authors wish to express their thanks to the Bundesministerium für Forschung und Technologie for having supported this work and the Fonds der Chemischen Industrie for a financial grant. We also wish to thank the Fraunhofer Gesellschaft für Angewandte Forschung, München, for administrative services. Furthermore, we cordially thank Mrs. Dr. Kubach, Gmelin-Institute, for helpful hints especially in literature concerning radium.

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