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DETERMINATION OF THE RADIOACTIVITY OF THE QUINCY GRANITE BY THE PHOTOGRAPHIC METHOD

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ABSTRACT. Approximate uranium and thorium contents of powdered Quincy (Mass.) granite have been determined with nuclear photographic emulsions. Compared with previous results, obtained by different methods, the new values have a wide range, because the radioactive elements are concentrated in distinct centers scattered haphazardly through the rock. The radioactive centers have been localized in thin sections by the emulsion techniques; they are found chiefly in sphene and ilmenite associated with riebeckite. The nuclear emulsion technique, which delineates the distribution of radioactive atoms in various accessory minerals may be useful in the study of the problem of geologic age of rocks.

INTRODUCTION

The Quincy granite has been studied by many workers, and the concentrations of radium, uranium, thorium, potassium and helium are given in various publications, particularly in the important work of Evans and Goodman (1941) on the radioactivity of rocks and in an article by Keevil (1938). Keevil's average values for the whole rock are given below:

Ra x 10^{12} g/g	U x 10^6 g/g	Th x 10^6 g/g	He x 10^5 cm^3/g	Th/U	K x 10^2 g/g
0.98	2.78	12.4	4.33	4.5	3.8

The potassium concentration was also given in an article by Hess (1947). This value agrees with that reported in a chemical analysis of the Quincy granite quoted by Osann (1916).

DETERMINATION OF URANIUM AND THORIUM CONCENTRATIONS IN POWDERED SAMPLES OF THE QUINCY GRANITE BY A PHOTOGRAPHIC METHOD

During the beginnings of our work on the photographic method applied to the study of uranium and thorium concentrations in powdered rock by the technique given by Madame Joliot-Curie (1946) and by Yagoda (1949) it seemed interesting to us to compare our results to those found by older and entirely different methods. The Quincy granite was ideally suited for this purpose. Professor P. M. Hurley of the Massachusetts Institute of Technology has kindly sent us a sample of this rock, for which we are deeply grateful.

Of our various analyses we list only those of two separate powdered samples:

	U x 10^6 g/g	Th x 10^6 g/g	Th/U	Ra x 10^{12} g/g
Powder 1 plate no. 311	5.1	18.7	3.7	1.7
Powder 2 plate no. 325	2.1	10.4	4.9	0.7

We believe the differences found in the concentrations of the elements are not due to experimental errors. One can see that the thorium-uranium ratio varies only within acceptable limits, but the concentrations of uranium and thorium vary simultaneously by almost a factor of two.

The study of the localization of radioactive centers of which we shall speak later leads us to believe that the differences in concentration are due

*We are indebted to H. Faul for this translation of our paper.

to the irregular distribution of radioactive centers in a mass whose radioactivity is low in comparison. According to our study the active surface is only 1.6 percent of the total surface. We shall see that the atoms of uranium and thorium are contained in very small crystals, the distribution of which is certainly far from homogeneous, so that we should not be surprised at the diversity of results obtained on different powdered samples.

Keevil (1938) gives the results of measurements on eight different samples of Quincy granite. These results show great variation as follows: 2.75 to 5.31 x 10^{-5} cm^3/g for helium, 0.77 to 1.28 x 10^{-12} g/g for radium, 5.9 to 12.4 x 10^{-6} g/g for thorium.

It is true that the same author (Keevil et al., 1943) later attempted to establish a law of distribution of radioactivity in batholiths. He observes a tendency for the radioactivity to decrease from the border toward the center. However, speaking of the granodiorite massif of Bourlamarque, he adds that the variation along a profile does not proceed gradually from specimen to specimen. Actually, two high values found near the center were due to an abnormally strong local concentration of zircon.

The relation between the radioactivity and the presence of certain accessory minerals cannot be escaped. We can hardly hope to find a perfect agreement between different specimens of a granite considering the fact that the crystals which contain the radiaoctive elements are irregularly distributed in the rock. One need only consider the extremely small size of the radioactive centers and their haphazard distribution in the relatively inert mass of the rock.

In the evaluation of the concentration of uranium and thorium in powdered samples of the Quincy granite we have considered the background of the Ilford plates. We have previously called attention to the importance of this factor (Hée et al., 1951). The background was determined by running a blank plate. In this case the background amounts to 30 to 40 percent of the measured radioactivity of the powders. This correction is of the same order of magnitude as that of the leakage current of a quadrant electrometer with which one measures the weak ionization currents due to the radioactivity of such rocks.

LOCALIZATION OF RADIOACTIVE CENTERS IN THE QUINCY GRANITE

We have used first the shadowing method previously described by one of us (Hée, 1948) for the study of opaque parts of petrographic thin sections. Afterward, the photographic emulsion (Ilford C2) was poured directly on the uncovered thin sections, following the suggestion of Ford (1951). This procedure is rapid and the precision of localizing radioactive centers in the transparent areas is perfect. In the opaque areas the tracks can be discerned along the borders, but it is prudent to study the thin section according to the shadowing method (Hée, 1948) as well, before covering it with a photographic emulsion.

Unfortunately, the presence of the photographic emulsion on the thin section adds to the difficulties of mineralogic determination of the radioactive centers, which frequently are extremely small. Nevertheless, with some experience, a good mineralogist becomes used to viewing the rock through the additional layer of photographic emulsion. Where it is absolutely impossible to recognize the minerals through the emulsion, one can carefully remove the emulsion after meticulously fixing the position of the radioactive centers. This orientation is fairly easy in the granite because of the very definite outlines of grains of major minerals. Wherever necessary, one can refer to photomicrographs made before the emulsion was removed.

Two thin sections were studied, numbers 36 and 28. The first was investigated over part of its surface and the second over its entire surface. The latter was, furthermore, especially examined from the point of view of possible contamination. Our emulsion was poured directly onto the section, thus minimizing the background. Table 1 shows the distribution of alpha tracks in these thin sections. In the various columns we have shown the following data:

Column 1. Exposure time.

Column 2. The number of important radioactive centers found on the examined area S of the thin section.

Column 3. The number of alpha particles emitted by the centers on each slide.

Column 4. The approximate surface S_c occupied by these centers.

Column 5. The examined area S of the thin section.

Column 6. The number of tracks on the area S-S_c.

Column 7. The average number of alpha tracks per centimeter square of S-S_c.

Column 8. The average number of tracks per centimeter square of the emulsion area exposed only to the glass backing of the thin section.

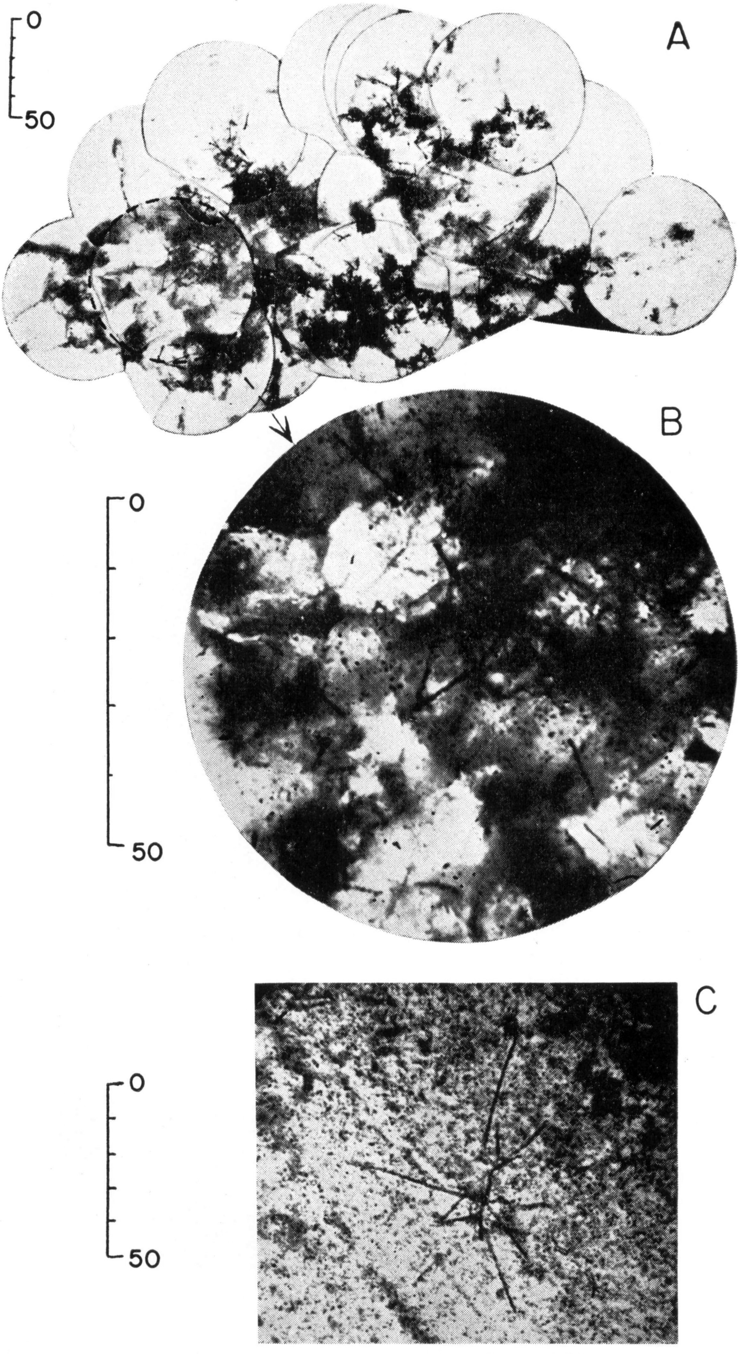
Column 9. The average number of tracks per square centimeter of emulsion on a blank slide.

TABLE 1

	1		2	3	4	5	6	7	8	9
	Days	Hours			S_c mm²	S mm²				
Number 28 Emulsion 396	25	21	53	877	2.89	189	449	241	139	
Number 36 Emulsion 503	19	17	9	409	1.065	186				
Blank Emulsion 340	13	0								98

If we admit a proportional relation between the number of background tracks and the time of exposure it becomes clear that the edges of section 28 are not particularly contaminated. The number of tracks seen in this part corresponds to the background on the blank. Furthermore, the number of tracks per square centimeter of surface S-S_c is 241 minus 139 equals 102, or of the order of magnitude one would expect from the radioactivity of the glass and the emulsion itself. Area S-S_c is occupied by feldspar, quartz, and ferromagnesian minerals. If we consider the ratio of the number of disintegrating atoms per unit area of the active centers to the number of disintegrating atoms per unit area of the other parts of the section, we find

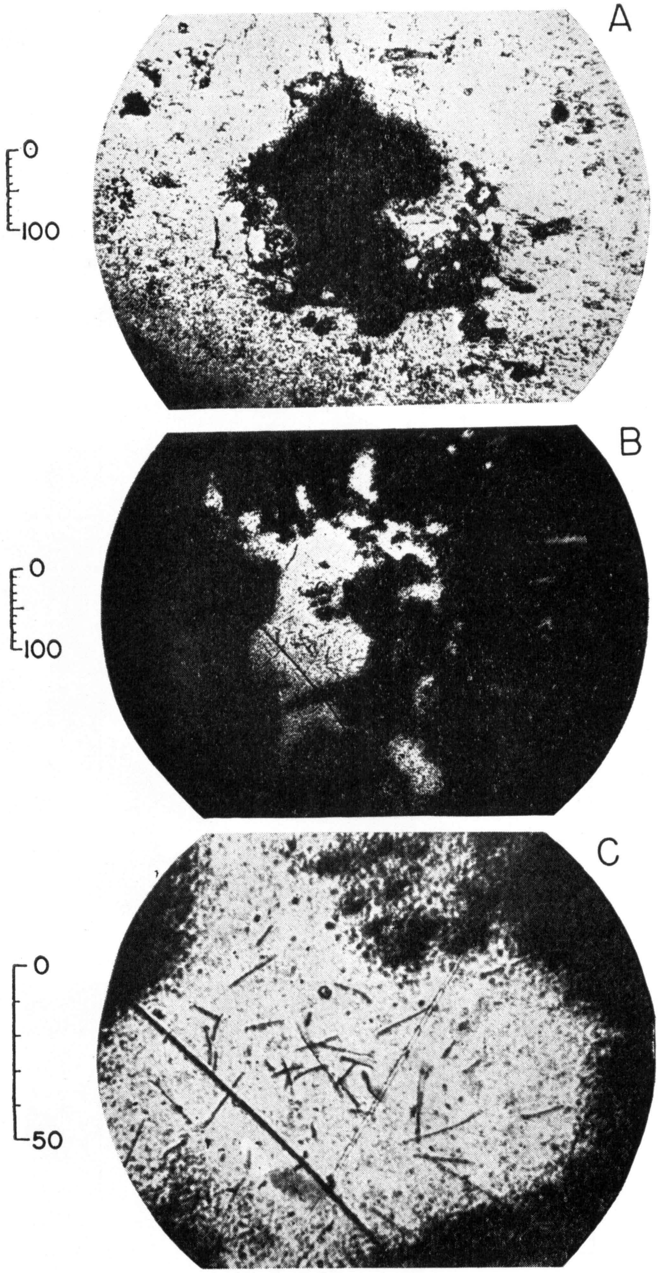
PLATE 1



Microphotographs of Quincy granite, Quincy, Mass.
Thin section 28, emulsion 396. Scales are in microns.

A. Center 4, large crystal of sphene.
B. Part of center 4, enlarged, showing α-particle tracks.
C. Center 149, small crystal of sphene?, showing radiating α-particle tracks.

PLATE 2



Microphotographs of Quincy granite, Quincy, Mass. Thin section 28, emulsion 371, center 67, ilmenite. Scales are in microns.
A. Positive print of thin section.
B. Ilford plate with α-particle tracks.
C. Part of same enlarged.

30,300/102 or about 300. This ratio shows how highly centralized in extremely small areas the radioactive atoms are, with the exception of certain parts of biotite near radioactive centers which seem to show an activity slightly higher than the background.

Picciotto (1950) in his study of the granite of Blanc Lake in the Vosges Mountains, Coppens (1950) in his study of the radioactivity of rocks, and ourselves (Hée, 1948, 1951) have already mentioned this segregation of radioactive atoms in rocks. Larsen and Keevil (1942) in their study of gravity separates of constituent minerals, have shown that in the Lakeview tonalite, Riverside County, California radioactive elements are concentrated in apatite, zircon and sphene.

SUMMARY OF THE PRINCIPAL RADIOACTIVE CENTERS FOUND IN TWO THIN SECTIONS OF THE QUINCY GRANITE

All the alpha tracks were measured with the aid of a microscope. The point of emission on the surface of the thin section is marked by a number. Table 2 lists the principal radioactive centers with their approximate dimensions, the number of alpha tracks, and the mineralogic identification of the centers. The exposure time is shown in table 1.

In summary, the active centers are found in sphene and in ilmenite located in the immediate vicinity of or even within riebeckite. The last mineral itself shows alpha tracks only rarely. The roughly estimated length of the tracks (without the establishment of histograms) shows that we are dealing with atoms that belong to the uranium and thorium families.

We should add that in both of these thin sections we have found only a single radioactive center which corresponds to a well-developed crystal. The crystal is brown, deeply colored, birefringent, and strongly pleochroic in dark brown and black, alpha equals zero. The strong natural coloring does not permit estimating the birefringence hues nor the sign of elongation. The mineralogic determination could not be made exactly, but we are dealing with a mineral similar to rutile of the variety nigrine, ilmeno-rutile, a colored variety of sphene (titanite), or allanite. This center does not appear in the list given in table 2 because it occurs in a region of thin section no. 36 which was not methodically investigated.

REMARKS ON THE MAGNETIC PROPERTIES OF THE QUINCY GRANITE

The chemical analysis given by Osann (1916) shows the presence of titanium which we find in sphene and in ilmenite. We have again shown what we had already mentioned (Hée et al., 1951); namely, that a rock of relatively moderate susceptibility does not necessarily contain magnetite but more likely ilmenite as the paramagnetic material. The susceptibility of the Quincy granite is of the order of 36 x 10^{-6}, analogous to that of the Raon-l' Etape granite in the Vosges Mountains (25 x 10^{-6}).

TABLE 2

Section	Number of Center	Dimensions μ	Number of Alpha Tracks	Mineralogic Identification
Number 28 Emulsion 396	1	100 x 100	16	Sphene
	4	300 x 200	100	Sphene in colored and highly birefringent grains, leaving the possibility that it may be a different mineral (see pl. 1-A and B).
	9	100 x 100	6	Sphene?
	14	300 x 300	7	Sphene and riebeckite
	16	200 x 200	9	Intergrowth of sphene, riebeckite, and ilmenite?
	67	1000 x 1000	150	Ilmenite?, opaque mass with clear and ilmenite?
	73	80 x 150	7	Sphene, ilmenite
	121	50 x 70	7	Sphene, ilmenite
	135	100 x 100	35	Three grains which are similar to number 4 and are difficult to identify because of the surrounding riebeckite.
	147	50 x 20	7	Sphene in riebeckite
	149	35 x 15	10	Sphene? See pl. 1-C.
	152	10 x 10	9	Sphene
	155	80 x 60	22	Sphene in riebeckite
	180	150 x 100	>100	Opaque mass like number 67, ilmenite?
	196	450 x 250	107	Sphene? Masked by a rust-colored mineral (limonite)
	240	50 x 50	11	Sphene
	250	15 x 15	12	Sphene
	258	100 x 20	20	Sphene?
	278		35	Ilmenite
	283	50 x 50	16	Sphene, riebeckite, ilmenite
Number 36 Emulsion 503	17	500 x 500	8	Sphene, locally sprinkled with ilmenite and all in a riebeckite crystal.
	25	200 x 40	12	Sphene, ilmenite
	27	100 x 80	27	Sphene
	52	200 x 150	25	Riebeckite according to the birefringent colors, masked by an undetermined mineral.
	60	650 x 200	38	Intergrowth of sphene, ilmenite, and riebeckite
	64	150 x 70 120 x 100 150 x 100 30 x 50	300	Four sphenes

COMPARISON WITH N. B. KEEVIL'S RESULTS ON THE QUINCY GRANITE

As we have already mentioned above, the work of Keevil showed about 10 years ago that radioactive atoms tend to be concentrated in certain accessory minerals. The photographic method confirms these early results as far as granites in general are concerned. The radioactive concentrations are found in zircon, sphene, magnetite, ilmenite, and apatite. However, Keevil et al. (1943) report mineralogic studies of two specimens of Quincy granite where they mention minerals which we have not encountered in our thin sections, for example, epidote, zircon, and needles of apatite. Like ourselves they mention the presence of numerous sphenes, but they have not been able to establish a direct relation between the presence of sphenes and total radioactivity in the Quincy granite. We are in disagreement on this point and we find it difficult to explain this divergence of opinion. The results of these early American researches are on the whole very precise, and the photographic method confirms them in all other respects.

CONCLUSIONS

The photographic study of powdered samples of Quincy granite has led to two principal results:

(1) The variations in uranium and thorium concentration found in photographic studies of powdered rocks are about the same as those one obtains by other methods. The study of the length and number of alpha tracks in photographic emulsion is laborious. However, this work is less difficult than the various operations required for the determination of radon and thoron by the classic methods. The apparatus described by Evans and Goodman (1941) appears to be the only one that is more rapid. Whatever method one uses, the determination of concentration of uranium and thorium in the total mass of a rock cannot be made with any reasonable accuracy. The indubitable establishment of this fact by the photographic method may lead to a new approach to the interpretation of helium age determinations on rocks.

(2) The photographic method is perfectly suited to the study of radioactive centers in rocks, particularly when one uses emulsions that have been deposited directly on the thin section and/or on the microscopic crystals which contain radioactive atoms. The small radioactive minerals can be separated from the large mass of relatively inactive material. In our current work we are proceeding in this general direction.

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