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THE DISTRIBUTION OF RADIOACTIVITY IN THE MAJOR ROCK FORMING MINERALS

BY

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WITH 19 FIGURES IN THE TEXT

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CONTENTS

Abstract.

The distribution of uranium (as determined by the nuclear emulsion method) and radioactivity is traced in the felspar, pyroxene, and quartz of the differentiated Skaergaard intrusion. It is characteristic that the radioactivity from the micro (and macro) inclusions in the major minerals progressively increases in the differentiated sequence towards the later members. The activity from inclusions is generally greater than that from the host mineral.

The Skaergaard intrusion is compared to various granites, granophyres, and basalts. The heterogenous distribution of radioactivity, related to the distribution of the accessory minerals, is shown by the Standard Granite G1.

In all the rocks examined the uranium, and radioactivity, even at the low levels encountered, always showed a characteristic pattern.

INTRODUCTION

Over half a century has passed since the first work was carried out on the distribution of radioactive materials in rocks. STRUTT (1905) was one of the first to observe that there was a concentration of radioactive constituents in the acid rocks. PIGGOT (1933) showed that in the case of the Hawaiian lavas the most radioactive were those poorest in silica; the most siliceous alkaline lava of Hualalai contained no more radium than the basalts of Kilauea. The concentration of the radioactive elements in accessory minerals such as apatite, zircon and sphene has been described by LARSEN (1942).

In acid rocks the accessory minerals in particular show a variable distribution. A theoretical approach to the distribution of accessory elements in pegmatites has been made by HOLLAND (1949). MOORHOUSE (1956) concludes that the accessory minerals were some of the last minerals to crystallise out rather than the first. The relative times of formation of of the accessory minerals depend upon many physical chemical factors, and must be considered individually for different rock types.

Excellent discussions on the distribution of uranium in igneous rocks, and the origin of ore deposits are given by LARSEN (1954) and MCKELVEY (1955). Because of the low concentration of uranium in basic magmas, and the large size of the quadrivalent uranium ion, uranium neither precipitates as a separate uranium mineral nor does it substitute isomorphously in the major minerals. In acid magmas uranium concentrates in the residual liquids and is then precipitated as a minor constituent in the accessory minerals in which it is in isomorphus substitution.

In some of the earlier work by JOLY (1909), HIRSCHI (1924—27), PIGGOT (1931), ROSNER (1933), URRY (1933), EVANS (1935), KEEVIL (1938 a, b, c), and LARSEN (1942) et. al. it has been shown that the radium content of rocks increases with silica and potassium. LARSEN (1942) describes the distribution of helium and radium in the minerals of the Lakeview tonalite. DAVIS (1947, 1949) applied the radium method to a study of the distribution of radium in ultramafic rocks. The radium is concentrated in late stage minerals such as talc, tremolite and kam-

mererite. The hydrous serpentines contain three times more radium than the anhydrous types. The distribution of uranium and thorium in some volcanic rocks has been described by ADAMS (1954, 1955) together with the relationship of uranium and potassium (as shown by LARSEN (1947). The variation of the uranium content of petrographically equivalent rocks is related by NEUERBURG (1955) to the distribution of the accessory minerals. Similar variations in the radium content of similar rocks have been described by EVANS (1935) and SOLOVIEV (1936), while EVANS (1941) notes no difference between a related plutonic and volcanic suite of rocks. LARSEN (1955) considers that the major minerals contain most of the uranium in an igneous rock. The uranium content of the quartz and felspar is approximately 50 % of that present in the rock. The enrichment of uranium in the mafic minerals in any rock series is in the youngest members. A study of the distribution of uranium and radioactivity in the differentiated Skaergaard intrusion by HAMILTON (1957, 1959) demonstrated the variation in these rocks. A substantial part of the uranium in an igneous rock is late and is readily leachable. The heterogeneous distribution of uranium and lead in a zircon (ROUBALT 1958) and of radioactivity in accessory minerals (HAMILTON 1959) show that the incorporation of uranium and thorium in these minerals does not follow a simple pattern.

The radioactivity of the rocks of the S. California batholith (LARSEN 1947) shows an increase from 0.3 alphas/mg/hr. in the gabbros to 2.0 alphas/mg/hr. in the granites. Similarly the activity from the felspar ranges from 0.34 alphas/mg/hr. (An_{93}) to 1.33 alphas/mg/hr. (An_{55}). Although the amount of radioactivity increases with potassium the potassium minerals have a low activity. SASTRY (1957) has shown that the radioactivity of some charnockites is associated with the alkali felspar; but with the mafic minerals in hybrid rocks and gneisses. While the uranium content of most rocks is less than 10 ppmU, the amount depends upon the past history of the rock. Some Black Forest gneisses (HUSMANN 1956) contain 5.5 ppmU, and associated anatectic and magmatic rocks contain 7.3 and 15 ppmU respectively.

The ratio of thorium to uranium is generally quoted as being ca. 3. This value cannot be assumed in geochemical considerations as it often varies considerably. HURLEY (1957) has shown that accessory minerals which also occur as later minerals in pegmatites have differing ratios.

In addition to obtaining data on the uranium and thorium content of rocks and minerals the use of nuclear emulsion techniques makes a study of the actual distribution of radioactivity possible. Some of the first work in this field was carried out by KINOSHITAS (1910), ALEXANDROV (1927), STEP (1940). BARANOV (1935) used thick emulsions in a study of the distribution of the radioactive elements. EVANS (1934, 1935, 1941),

CURIE (1946), COPPENS (1949), PICCIOTTO (1949) and FARAGGI (1950) studied the alpha particle emission from thick and thin sources and the preparation and development of nuclear emulsions. A general survey of the method is given by BOWIE (1954).

Compared with the amount of work done on the determination of uranium and thorium in rocks the nuclear emulsion method as a diagnostic tool is in its infancy. To be of use the method must first of all be applied to genetically related rock sequences. PICCIOTTO (1949, 1950, 1952), POOLE (1949), MERLIN (1957), DEUTSCH (1958), and HEE (1954) have applied the method to a variety of rocks and established the main trends. The author (1957, 1959) has applied the method to the Skaergaard rock sequence, and to a preliminary survey of basalts.

This paper deals mainly with the specific alpha particle activity of the major rock forming minerals (in particular felspar, pyroxene and quartz), and the accessory minerals. The felspar being the commonest of rock forming minerals are of particular interest since the changes in radioactivity may be traced through a variety of rocks.

In this work the thin sections of rock were covered with gel emulsion and the alpha tracks observed by means of a binocular microscope at magnifications of 500, and 1.000. For the latter a Leitz 53 objective was used and × 16 wide view occulars. This combination made observations easier and reduced eye strain.

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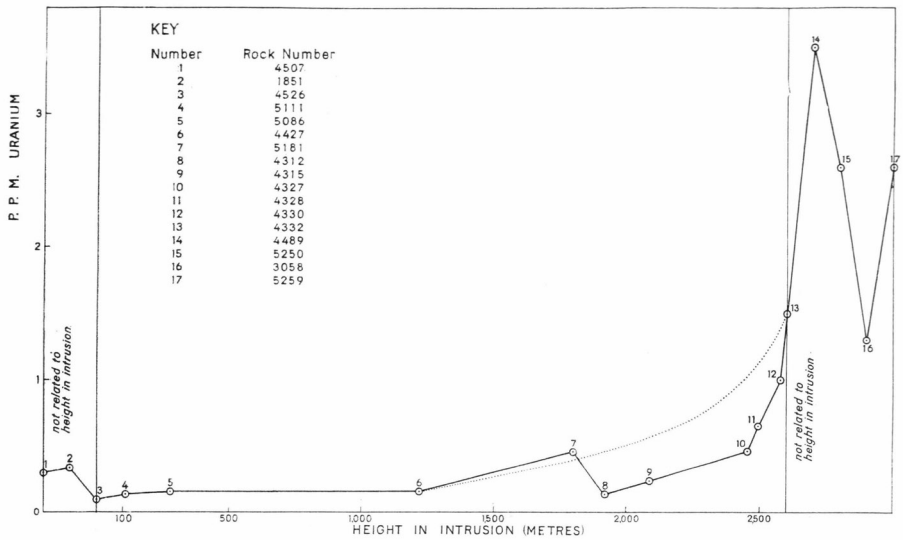


Fig. 1. Plot of the distribution of uranium against height in the Skærgaard intrusion. The dotted line represents the uranium contents corrected for weathering.

Key. 1. Chilled marginal gabbro. 2. Perpendicular felspar rock. 3. Gabbro picrite. 4, 5. Hypersthene olivine gabbros. 6. Middle gabbro. 7. Hortonolite ferrogabbro. 8, 9, 10, 11. Ferrogsbbros. 12. Basic hedenbergite granophyre. 13. Basic granophyre. 14, 15. Transgressive granophyres. 16, 17. Acid granophyres.

1. THE DISTRIBUTION OF RADIOACTIVITY IN THE SKAERGAARD INTRUSION

The distribution of the trace elements in the Skaergaard intrusion has been described by WAGER and MITCHELL (1951) and the distribution of uranium and radioactivity has been described by the present author (1958). Uranium (and thorium) were clearly concentrated in the residual liquid, and the resultant granophyres are the most radioactive rocks of the differentiated sequence. The distribution of uranium in the intrusion is given in Fig. 1, the dotted line represents the expected uranium content of those rocks which are suspected of being weathered. The distribution of radioactivity in the major minerals of the intrusion is given in Fig. 2.

The analyses were made by nuclear emulsion techniques, and show that the radioactivity of the essential minerals is almost constant. The slight variations shown are near the limit of error covered by analyses and sampling. The total alpha activity from the mineral "a", including that from inclusions cracks and alteration, is compared to the activity contributed to by the pure mineral "b". In the rocks studied the activity

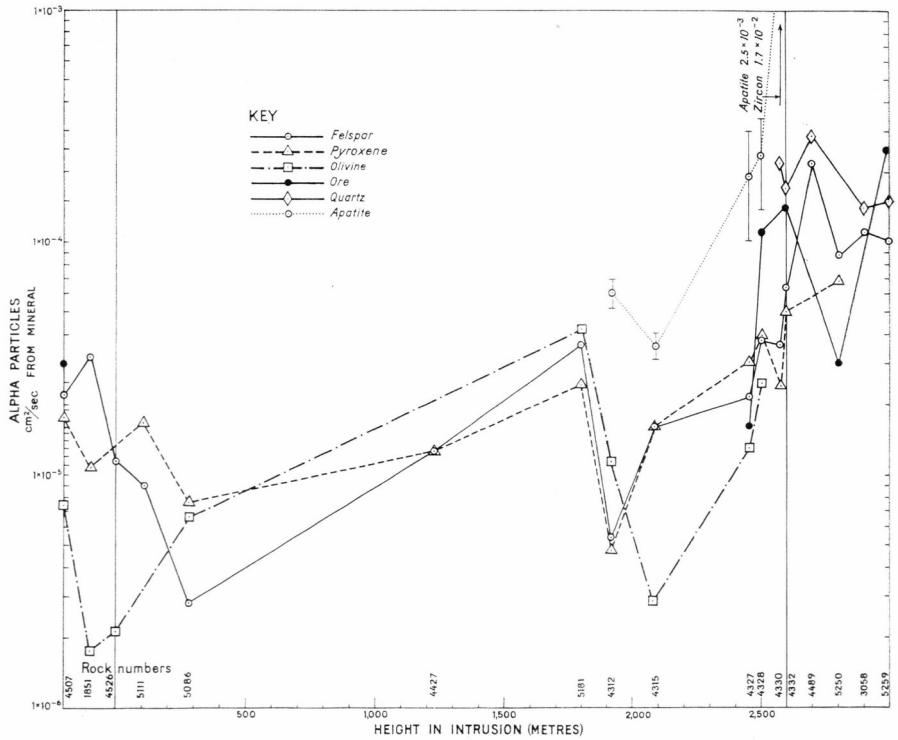


Fig. 2. Plot of the alpha particle activity from the individual minerals against height in the intrusion. [Rock numbers see Fig. 1].

from cracks and alteration was generally very small. The word inclusion is used to describe any foreign body present in a mineral. These may be other minerals or small indefinite spots.

a) Felspar.

In Table 1 the distribution of radioactivity in the felspar is given and the appropriate uranium concentrations shown assuming that the Th/U ratio is 3. The ratio a/b in the gabbros and ferrogabbros is never less than one nor greater than two. This indicates that although there is no significant increase in radioactivity from either the felspar or from inclusions within it, is a tendency for the inclusions to be slightly more active. In the Skaergaard rocks a linear relationship exists between the alpha particle activity as deduced from nuclear emulsions and the total uranium content as determined by neutron activation and fluorimetry. This is taken as an indication that uranium and thorium did not follow different lines during differentiation. The maximum enrichment of uranium and thorium is in the basic, acid and transgressive granophyres.

Table 1. The distribution of uranium and radioactivity in felspars from the Skaergaard intrusion.

Sequence	Rock number	Rock type	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (b)	Uranium content ppm. U.	Ratio $\frac{(a)}{(b)}$
a) Marginal Series	E.G. 4507	Chilled marginal gabbro	2.2	0.09	1.1	0.05	1.9
	E.G. 4526	Gabbro perite	1.1	0.04	1.0	0.04	1.1
	E.G. 1851	Perpendicular felspar rock.	3.2	0.13	1.3	0.05	2.5
b) Lower Gabbros	E.G. 5111	Hypersthene olivine gabbro	0.9	0.04	0.8	0.04	1.1
	E.G. 5086	—	0.3	0.02	0.3	0.01	1.2
	E.G. 4427	Middle gabbro	1.2	0.05	1.0	0.04	1.2
c) Upper Gabbros	E.G. 5181	Ferrogabbro	3.6	0.15	2.3	0.09	1.6
	E.G. 4312	—	0.5	0.02	0.5	0.02	1.1
	E.G. 4315	—	1.6	0.07	1.1	0.04	1.0
	E.G. 4327	—	2.2	0.09	1.0	0.04	2.1
	E.G. 4328	—	3.8	0.16	2.4	0.10	1.3
d) Granophyres	E.G. 4330	Basic Hedenbergite granophyre	3.6	0.15	0.6	0.03	6.0
	E.G. 4332	Basic granophyre	6.4	0.22	0.6	0.03	10.0
	E.G. 5250	Transgressive granophyre	8.7	0.31	0.6	0.03	14.5
	E.G. 5259	Acid granophyre	2.7	0.11	1.5	0.06	1.8

[1]) Uranium determined by the nuclear emulsion method. The results are quoted to the second decimal place only. (a) The total activity from the felspar. (b) The activity from the felspar.

b) Pyroxene.

The distribution of radioactivity in the pyroxenes of the Skaergaard intrusion is shown in Table 2. The ratio of activity from the pyroxene to that from the inclusions is never less than one, and shows a slight increase in the later rocks derived from the residual liquids. There is no great increase in activity from the inclusions of the later stage pyroxenes as was found with the felspar. The ferro-hortonolite ferrogabbro E.G. 4312—4315 contain up to 6 % of the primary precipitate apatite which is enriched in uranium and thorium. The a/b ratio of these rocks is only slightly greater than that of the hortonolite ferrogabbro containing

Table 2. The distribution of uranium and radioactivity in the pyroxenes from the Skaergaard intrusion.

Sequence	Rock number	Rock type	$\alpha/cm^2/sec \times 10^{-5}$ Pyroxene (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Pyroxene (b)	Uranium content ppm. U.	Ratio $\frac{(a)}{(b)}$
a) Marginal Series	E.G. 4507	Chilled margin gabbro	1.78	0.06	0.74	0.03	2.4
	E.G. 1851	Perpendicular Felspar Rock.	1.06	0.04	0.74	0.03	1.4
b) Lower Gabbros	E.G. 5086	Hypersthene divine gabbro	0.75	0.03	0.25	0.01	3.0
	E.G. 4427	Middle Gabbro	1.24	0.04	1.20	0.04	1.0
c) Upper Gabbros	E.G. 5181	Ferrogabbro	2.40	0.09	1.90	0.07	1.3
	E.G. 4312	—	0.47	0.02	0.27	0.01	1.7
	E.G. 4315	—	1.60	0.06	1.20	0.04	1.3
	E.G. 4327	—	3.05	0.11	0.96	0.04	3.1
	E.G. 4328	—	3.95	0.14	1.20	0.04	3.3
d) Granophyres	E.G. 4330	Basic Hedenbergite granophyre	2.40	0.09	0.55	0.02	4.4
	E.G. 4332	Basic granophyre	5.10	0.18	2.70	0.09	1.9
	E.G. 5250	Transgressive granophyre	6.40	0.23	1.50	0.05	4.3

practically no apatite. The fayalite ferrogabbros E.G. 4327—4328 contain small interlocking grains of pale green pyroxene formed by the inversion of an iron rich beta wollastonite. The pyroxene is cloudy and full of inclusions of iron ore. Most of the activity originates from areas of alteration in the pyroxene. It is significant that over 50 % of the emanation in these rocks comes from the pyroxene.

The ferrogabbro E.G. 4328 contains up to 0.52 % of sulphur (Wager 1958).

c) Quartz.

Only E.G. 4332, 5250, and 5259 contain sufficient quartz for analyses. If the rock contains only a granophyric mixture of quartz and felspar it can be hard to distinguish between them under the microscope. In the early ferrogabbros quartz-felspar intergrowths occur in intercrystal areas but they were always inactive. The Skaergaard results are given in Table 3 together with those from other acid rocks. There is a slight increase in the activity from inclusions over that present in pure quartz.

d) Accessory Minerals.

The maximum enrichment of radioactivity occurs in the accessory minerals. Apatite is the most common accessory mineral present, others present include epidote sphene, zircon, and secondary micaceous material. The accessory minerals in the gabbros, and ferrogabbros contribute less than 30 % of the total radioactivity from the rock. In the late stage acid granophyres amounts greater than 67 % of the activity comes from secondary minerals.

e) Intercrystal Activity.

It is difficult to evaluate the significance of the radioactivity from the intercrystal areas on account of its sporadic distribution. However, it would seem that it contributes between 1–14 % of the total activity from the rock with an average value at c. 4 %.

WAGER (1939) has shown that the chilled marginal gabbro contains xenoliths of gneiss derived from the country rock. They are present in the form of abundant patches and streaks of granophyric material. The work of HOLGATE (1954) and WAHL (1946) describe the effect of acid xenoliths in basic magma, and the effect of thermal diffusion—convection as a cause of magmatic differentiation. Apart from the marginal gneiss inclusions, the large gneiss rafts of Tinden and Basistoppen could have an influence on the composition of the late stage granophyres, above the level of the fayalite ferrogabbro. WAGER and MITCHELL (1951) consider the gray quartz gneiss xenoliths as fluid viscous masses surrounded by a partly crystallised basic material.

Extrapolation of the uranium content of the differentiated Skaergaard sequence (below the fayalite ferrogabbro) would indicate that the Hidden Layered Series (H.L.S.) had a uranium content lower than the hypersthene olivine gabbros that have been analysed. The H.L.S. are predictated to be enriched in sodium and aluminia, such that the plagioclase would have a composition of a sodium bytownite (An_{72} 66 % by weight) as compared to the olivine accumulate (Gabbro Picrite) with a calcic bytownite (An_{87} 23 % by weight) LARSEN (1947) has shown the increase of alpha particle activity with an increase in the amount of albite. The Skaergaard felspars show a similar increase. In the light of this evidence the H.L.S. may be relatively enriched in uranium compared to the lowest rocks exposed.

As the chilled marginal gabbro E.G. 4507 represent xenolith free material the average uranium content of c. 0.2 ppmU is that expected by taking a simple arithmetical mean of the differentiated Skaergaard sequence. In addition a value of 0.2 ppmU is common for olivine basalts.

Unfortunately the increase in the uranium content (if any) due to the acid xenoliths cannot be estimated with any certainty.

The effect of contamination because of the incorporation of country rock is probable in the case of the acid granophyres. The amount of contamination may be traced in the ratios of the alkalis Na_2O and K_2O. The alkali ratio of the gray gneiss (Mellemö) of 1.74, the granophyre inclusions in the layered series 1.79, the granophyre in the border group 4.75 may be compared to that of 2.43 of average gneiss and granophyre inclusion. The latter must only be approximate as it is not possible to obtain a true value for the bulk composition of the gneiss.

Of the trace elements (WAGER, MITCHELL 1951) zirconium and the rare earths may be used as tracer indicators for the variations in the amount of uranium and thorium. The values are given in Table 2 A.

Table 2 A.

Rock type	Zr. ppm.	Y. ppm.	La. ppm.
Gneiss	160	c. 30	c. 30
Granophyre inclusions	2500	350	280
Estimate of metamorphic gneiss	140	c. 30	c. 30
Inclusions in Western Border Group	500	200	300
	+ 1 %	200	300
	3000	300	300
	200	300	100
Inclusion in lower layered series	2000	800	500
Acid granophyre	850	250	240
Transgressive Hedenbergite granophyre	1400	350	80
Basic granophyre	250	180	c. 30
Fayalite ferrogabbro	25	70	0

The influx of sodium into the xenoliths might be accompanied by zirconium. It is doubtful if the uranium content of the granophyres represents uncontaminated material derived from the differentiated sequence. In comparison, the acid segregration of the Whin Sill (dolerite) does not show an enrichment factor comprable to the Skaergaard granophyres.

2. THE DISTRIBUTION OF RADIOACTIVITY IN SOME ACID IGNEOUS ROCKS

The distribution of radioactivity in the acid igneous rocks presents many problems. The sporadic distribution of the accessory minerals result in large sampling errors. In addition the distribution of uranium and thorium in granites and granophyres often depends upon the type and mode of emplacement of these rocks. In spite of these sources of error, the late stage Skaergaard granophyres, the granophyres and micro-

Table 3. The distribution of uranium and radioactivity in quartz from a selection of acid igneous rocks.

Locality or rock number	Rock type	$\alpha/cm^2/sec \times 10^{-5}$ Quartz (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Quartz (b)	Uranium content ppm. U.	Ratio $\frac{a}{b}$
G1. U.S.A.	Granite	3.80	0.15	0.75	0.03	5.1
Lac Blanc[1])	Granite	30.0	1.20	1.6	0.06	21.0
Elbe	Granite	64.0	2.60	45.0	1.90	1.4
Kasai	Granite	23.0	0.90	11.0	0.40	2.1
Adamello	Granodiorite	7.2	0.28	4.1	0.16	1.8
Skye Granophyres						
H 185	Granophyre	2.6	0.10	0.9	0.04	3.0
H 830	Granophyre	3.2	0.12	—	—	—
H 857	Granophyre	2.8	0.11	1.8	0.07	1.6
H 930	Microgranite	1.8	0.07	1.2	0.05	1.5
H 4036	Microgranite	5.0	0.20	2.5	0.10	2.0
Skaergaard Granophyres						
Eg 4332	Basic Granophyre	18.0	0.71	14.0	0.55	1.3
Eg 5250	Transgressive Granophyre	9.3	0.37	5.5	0.22	1.7
Eg 5259	Acid Granophyre	15.0	0.59	11.3	0.44	1.3

[1]) 71 % of the activity is in cracks in the quartz.

Table 4. The distribution of uranium and radioactivity in felspar from a selection of acid igneous rocks.

Locality or rock number	Rock type	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (b)	Uranium content ppm. U.	Ratio $\frac{a}{b}$
G1. U.S.A.	Granite	7.3	0.30	0.59	0.03	12.2
1. Lac Blanc	Granite	60.0	2.40	25.00	1.00	2.4
Elbe	Granite	42.0	1.70	9.70	0.40	4.3
2. Kasai	Granite	16.0	0.50	9.00	0.30	1.8
3. Adamello	Granodiorite	8.4	0.34	3.70	0.15	2.3
Skye Granophyres						
H 185	Granophyres G1.	0.86	0.04	0.43	0.02	2.0
H 830	—	2.90	0.12	2.60	0.18	1.1
H 857	—	3.40	0.14	1.47	0.06	2.3
H 930	Microgranite G2.	7.80	0.31	1.00	0.04	7.8
H 4036	—	14.60	0.59	1.70	0.07	8.6

1. Picciotto 1950. 2. Picciotto 1952. 3. Picciotto 1957.

granites of Skye, and the granites studied by Picciotto show striking similarities with the differentiated Skaergaard sequence in their activity distribution.

a) Felspar.

Although the actual alpha particle activity of the felspars is generally higher in the acid rocks the ratio of a/b is fairly constant. There is often an increase in activity coming from areas of alteration and from cracks in the felspar. The sequence of granophyres of Skye (G1-early granophyres, G2-later granophyres and microgranites) may be compared to the Skaergaard sequence. In particular the later G2 phase shows a similarity to the transgressive granophyres.

b) Quarz.

Although the activity from the inclusions in quartz is at least twice as great as that from the quartz itself the ratio a/b is comparable to that obtained from the felspars. High activity from cracks in quartz as was demonstrated in the Lac Blanc granite is not uncommon (Picciotto 1950).

c) Mafic Minerals.

The maximum enrichment of radioactivity in the major minerals occurs in the pyroxene, amphibole and micas. The intense natural colour

of some biotites and amphiboles makes it difficult to locate the actual source of an alpha particle track. The ratio a/b ranges from 2.5 to 10. The mafic minerals in granites often contain radioactive accessory minerals present as inclusions or concentrated near the crystal margins.

d) Accessory Minerals.

The irregular distribution of the accessory minerals makes it difficult to compare the amount of radioactivity that they produce. In the thin sections examined, the accessory minerals contributed over 30 % of the total activity from the rocks. Generally, however, the average is about 80 % of the total activity from the rock.

3. DISTRIBUTION OF RADIOACTIVITY IN SOME BASALTS AND RELATED ROCKS

Although only a few samples have been examined the distribution patterns are similar to those of the Skaergaard intrusion. The total uranium content of these rocks (assuming that the U/Th ratio is similar to that of the Skaergaard rocks) increases in the "acid differentiates", while the a/b ratio shows no large variation in felspar and pyroxene.

a) Whin Sill.

The Whin Sill forms an intrusive sheet in the Lower Carboniferous of Northern England. The general characters and petrology of the normal rock type of the Whin Sill, its compact marginal facies and its coarse pegmatitic segregations have been described by HARWOOD (1928) and TOMKIEFF (1928). The sill rocks can be divided into three main types;—

i) The fine grained basaltic selvages, occurring along the upper and lower contacts.

ii) Medium grained doleritec, forming the bulk of the sill.

iii) Coarse grained gabbroic type occurring as bands, segregation veins, and spheroids and always sharply separated from the normal rock.

In addition, there are local red acid segregations composed of turbid felspar and micropegmatite. These red vienlets are regarded as representing a pure acid differentiate of the quartz dolerite magma.

The uranium content of the various types analysed are-

i) Chilled Margin (1223) ..... .20 ppm U
ii) Medium grained rock (H 1575) ..... .25 ppm U
iii) Acid segregation (H 1575)..... .50 ppm U

(The uranium determinations were made by the emulsion method.)

The distribution patterns are given in Table 5. The acid differentiate shows features similar to those of the acid granophyre of Skaergaard (E.G. 3058, 5259) but not to those of the transgressive granophyres (E.G. 4489, 5250).

Table 5. The distribution of uranium and radioactivity in felspar and pyroxene from basalts.

Rock number Locality	Rock type	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Felspar (b)	Uranium content ppm. U.	Ratio $\frac{(a)}{(b)}$	$\alpha/cm^2/sec \times 10^{-5}$ Pyroxene (a)	Uranium content ppm. U.	$\alpha/cm^2/sec \times 10^{-5}$ Pyroxene (b)	Uranium content ppm. U.	Ratio $\frac{(a)}{(b)}$
1575 Whin Sill.	Dolerite	2.0	0.08	0.84	0.04	2.4	—	—	—	—	—
T 222 Hekla Iceland	Olivine basalt	1.9	0.08	0.87	0.04	2.2	1.7	0.06	0.85	0.03	2
E.G. 1093 Kap Dalton E. Greenland	Olivine basalt	1.3	0.05	0.65	0.03	2.0	0.62	0.02	0.37	0.01	1.7
E. G. 1112 Kap Daussy E. Greenland	Olivine basalt	1.9	0.08	1.2	0.05	1.6	0.96	0.03	0.43	0.02	2.2
E.G. 1057 Kap Dalton E. Greenland	Tholeiitic basalt	3.8	0.17	2.3	0.10	1.7	—	—	—	—	—
H 3616 Giants Causeway	Tholeiitic basalt	3.8	0.18	1.9	0.08	2.0	1.1	0.04	0.74	0.03	1.5

b) Olivine basalts.

The basalts T222, and E.G. 1093, containing .3 and .25 ppmU respectively, are comparable in uranium content to the chilled marginal gabbro of Skaergaard. The radioactivity from the felspar is slightly greater than that from the pyroxene, olivine and iron ore. WAGER (1939) describe E.G. 1112 as an olivine basalt, the thin section examined contained .8 percent of brown glass. This rock contains .32ppmU and the distribution pattern (Table 5) shows affinities to that of the tholeiitic basalt type.

c) Tholeiitic basalts.

The tholeiitic basalt E.G. 1057 is described by WAGER (1939) and H3616 by PATTERSON (1951). The uranium content of these two rocks is .66 and .52ppmU respectively. A characteristic feature of the tholeiitic basalts is the relatively high activity from the glass.

4. THE DISTRIBUTION OF RADIOACTIVITY IN THE WESTERLY RHODE ISLE GRANITE G1.

The Westerly Rhode isle granite (G1) is a fine grained leucocratic rock, and has been described by CHAYES (1950). In thin section it consists of quartz, microline, plagioclase (oligoclase), small amounts of biotite, muscovite, about 1 percent of opaque, and about 5 percent of non-opaque accessories. The non-opaque accessories observed in the particular thin sections examined included sphene, apatite, carbonate (mainly restricted to the plagioclase) and zircon.

Table 6. The distribution of radioactivity and uranium in G1. No. 1, 2, 3.

Mineral	Total Alpha Particle Activity			% Activity from Rock			% Activity from Mineral			Alpha Particle Activity $\times 10^{-5}$ $\alpha/cm^2/sec$ 3			Average uranium content $\times 10^{-6}$ g/g
	1	2	3	1	2	3	1	2	3	1	2	3	
Felspar	410	249	309	30	19.24	11.95	100	100	100	9.73	5.54	6.48	0.27
Felspar	68	2	31	5.0	0.16	1.19	16.6	1.0	10.0	1.60	0.06	0.65	0.03
Inclusions	280	242	250	20.5	18.69	9.68	68.0	97.0	81.0				
Cracks	62	5	28	4.5	0.39	1.08	15.4	2.0	9.0				
Quartz	94	68	48	6.9	5.25	1.86	100	100	100	4.83	3.76	2.89	0.16
Quartz	29	2	10	2.1	0.16	0.39	30.0	3.0	21.0	1.45	0.11	0.61	0.03
Inclusions	50	59	26	3.7	4.55	1.01	54.0	86.7	54.0				
Cracks	15	7	12	1.1	0.54	0.46	16.0	10.3	25.0				
Biotite	219	77	197	16.0	5.9	7.62	100	100	100	73.8	39.0	98.2	0.28
Muscovite	4	8	27	0.4	0.62	1.01	100	100	100	5.93	11.87	43.0	
Apatite	4	0	10	0.4	0	0.39	100	100	100				
Zircon?	61	6	4	4.5	0.47	0.15	100	100	100				
Accessory A + ORE	50	0	37	3.7	0	1.92	100	100	100				
Intercrystal	74	67	69	5.4	5.18	2.67	100	100	100				
Accessory Intercrystal	34	259	0	2.5	20.10	0	100	100	100				
Accessory B	220	117	59	16.0	9.05	2.28	100	100	100				
Acessory C	194	443	1789	14.2	34.24	69.18	100	100	100				
Total Area sq cms.	2.6	2.6	2.2										

Table 7. — 7 a Modal analyses of G1. (Volume %).
7 b Modal analyse of W1. (Volume %).

Mode. Vol %	Slide No.			*U.S.G.S. Bull. No. 980
	1	2	3	
Quartz	29.4	27.3	28.9	27.5
Felspar[1])	63.6	67.9	66.0	66.8
Biotite	4.4	2.9	3.6	3.2
Muscovite	1.0	1.0	1.1	1.3
Opaque Accessories	1.1	0.5	0.1	0.8
Non-paque Accessories	1.4	1.4	1.3	1.4
Length of Count	1,492	1,535	1,432	1,502

[1]) Includes K-Felspar and Plagioclase

Mode. Vol %	Slide No.		*U.S.G.S. Bull. No. 980
	1	2	
Felspar and Quartz[1])	50.6	48.3	49.8
Augite	44.5	45.0	45.0
Biotite	2.8	3.4	1.8
Opaque Accessories	1.8	2.8	3.3
Non-opaque Accessories	0.3	0.5	0.2
Length of Count	1,480	1,475	1,345

[1]) Includes K-Felspar, Plagioclase, Quartz (1.8 %)

*) Modal. Analyses of G1, W1, and the average values of U.S.G.S. Bull. No. 980. (Chayes 1950).

Because of the heterogeneous distribution of accessory minerals in granites three sections of the rock were cut in sequence at a spacing of 2 mm apart from each other.

The thin sections of G1 were covered with Ilford G Special gel emulsion and exposed for a period of thirty days at minus 0°C. The tracks were then observed under a magnification of $\times 1.000$. Some of the possible sources of error in the nuclear emulsion method are.

a) The loss of members of the uranium and thorium series during the preparation of the thin sections. This in particular applies to the radioactive elements present on hydrated iron oxide films, and to soft or water soluble minerals.

b) The difficulty in determining whether or not an alpha particle is coming from the body of the mineral or from an inclusion. As the diameter of the inclusion approaches the diameter of the end silver grain of the alpha particle track the uncertainty increases.

c) The loss of minerals or parts of minerals during the preparation of the thin sections.

Causes of the variations in radioactive data have been described by KEEVIL (1943) the most important of which is probably inadequate sampling. The analyses of the radioactivity in the three sections examined are given in Table 6. The modal analyses given in Table 7 agrees well with the average values given by CHAYES (1951).

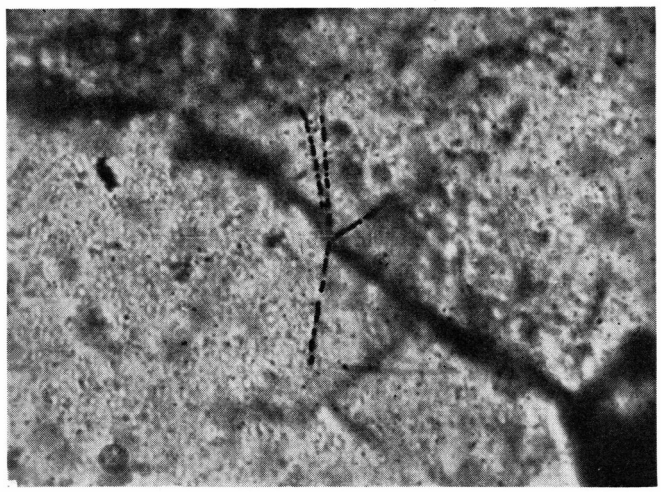


Fig. 3. Intercrystal activity G.I.

Fig. 4. Intercrystal accessory mineral G.I.

a) Activity from the major minerals.

Ten to twenty percent of the total radioactivity of the rock originates from inclusions within the felspar. Activity from cracks within the felspar appears to be quite sporadic. Assuming a U/Th ratio of 1/3 for the felspar (U/Th of the rock is 1/14) the alpha particle activity corresponds to a uranium content of less than 10^{-7} ppmU.

The radioactivity from quartz originates from inclusions within the quartz. The nature of these inclusions are unknown.

The natural colour of the biotite makes it very difficult to be certain whether the radioactivity originates from the minerals or from inclusions within it. Apparently practically all the activity comes from inclusions within the mineral or from marginal areas where the biotite has been replaced by chlorite. An increase of radioactivity in chloritised biotite has been described by ROUBALT (1958). The activity amounts to between 5—16 percent of the total radioactivity from the rock. The uranium content is in the order of 2—4 ppmU. Most of the radioactivity from muscovite comes from inclusions within the mineral.

b) Activity from the accessory minerals.

The non-opaque accessories constitute less than .5 percent of the total minerals by volume. Apatite larger than 10 microns (c axis) was generally inactive. Accessory No. A is probably apatite and occurs as small prismatic crystals less than 50 microns long adjacent to ore minerals and biotite. Accessory No. B and C appears to be sphene and occurs as clean crystals but sometimes as spongny aggregates. Type No. C, leucoxene is either formed as an alteration product of type No. A or occurs separately. Fluorite, and iron ore were inactive. The accessory minerals in the three rocks contribute between 40—80 percent of the total radioactivity of the respective rock.

Intercrystal radioactivity (Fig. 3) is defined as the activity that occurs along the junction planes of the minerals. The intercrystal activity is fairly constant. The intercrystal accessories (accessory minerals, Fig. 4, occuring between the major minerals) consisted of sphene or ferruginous secondary products.

The distribution pattern obtained from G1 is one fairly typical for granites. Of the inclusions in the major minerals those in the felspars are the most radioactive. Most of the activity comes from the accessory minerals.

5. THE DISTRIBUTION OF RADIOACTIVITY IN THE CENTERVILLE DIABASE W1.

The Centerville diabase (W1) described by CHAYES (1951) consists of augite, plagioclase (laboradorite), with smaller amounts of quartz, potash felspar, biotite, and opaque minerals. The modal analyses of W1 is given in Table 7b and the distribution of radioactivity in Table 8.

The inclusions in the felspars constitute the most radioactive component of the major minerals. The intercrystal activity is quite sporadic. Of the accessory minerals orthite is the most active. It is present in very small amounts with not more than one or two crystals present per slide.

The total uranium and thorium contents of G1, W1 (chemically determined) are given in Table 9.

Table 8. The distribution of radioactivity and uranium in W1. No. 1, 2.

Mineral	Total Alpha Particle Activity		% Activity From Rock		% Activity From Mineral		Alpha Particle Activity $\alpha/cm^2/sec \times 10^{-5}$ g/g.		Average Uranium Content $\times 10^{-6}$ g/g.
	1	2	1	2	1	2	1	2	
Felspar	101	62	34.89	60.20	100	100	3.08	2.14	0.18
Felspar	6	7	3.26	6.80	5.94	11.29	0.19	0.24	0.01
Inclusions	94	41	51.09	39.81	93.07	66.13			
Cracks	1	14	.54	13.59	.99	22.58			
Augite	7	12	3.80	11.65	100	100	2.43	4.45	0.12
Augite	0	3	0	2.91	0	25.0	0	1.11	0.04
Inclusions	5	2	2.72	1.94	41.4	16.6			
Cracks	2	7	1.08	6.80	28.6	58.4			
Biotite	8	2	4.35	1.98	100	100	4.41	1.11	0.02
Apatite	3	1	1.62	0.99	100	100	10.80	3.02	
? Orthite	18	8	9.78	7.80	100	100			
Intercrystal	47	18	25.56	17.50	100	100			

Table 9.

Uranium ppmU.

G-1	W-I	Method	Analyst
3.5, 3.6	.52, .54	Neutron activation	Hamilton 1957
3.2 (Average of 17)	.51 (Average of 13)	Fluorimetry	Hamilton 1957
3.9 (Average of 4)	.50 (Average of 3)	Gamma-ray Spectrometry	Hurley 1951
3.8 (Average of 46)	.52 (Average of 40)	Fluorimetry	Joslyn 1958

Thorium ppm. Th.

G-1	W-I	Method	Analyst
—	2.6	Chemical	Grimaldi 1957
45 (Average of 15)	(Average of 9)	Chemical	Grimaldi 1957
61	3.6	X-Ray Fluorescence	Hurley 1957
52 (Average of 14)	2.2 (Average of 5)	Chemical	Levine 1958
51 (Average of 14)	2.2 (Average of 5)	Chemical	Levine 1958
51 (Average of 4)	2.1 (Average of 4)	Chemical	Hamilton 1959

6. CONCLUSIONS

The uranium results calculated from nuclear emulsion data assume a uranium to thorium ratio of 1:3. It is not considered that this leads to large errors in the case of the major minerals. Variations in the U:Th ratio of rocks often vary because of the presence of the more radioactive accessory minerals such as apatite, orthite, zircon, sphene and others. The distribution of these is generally of a heterogenous nature.

1) The results derived from the analyses of the Skaergaard intrusion show an increase of uranium and thorium in the later more acid rocks. This conclusion is supported by the acid segregations in the Whin Sill, and the increase of radioactivity in areas of glass of tholeiitic basalts. The recent work on the differentiates of basaltic magma, (andesites, trachytes, rhyolites) as yet unpublished, further supports this.

2) In defining an inclusion in a mineral as a foreign body discernable under the microscope at high or low magnification, the inclusions always have a higher uranium content than the host (major) mineral. The difference is less pronounced in the more basic rocks. In a previous paper (HAMILTON 1959) the ratio of the percentage of the total radioactivity in the felspar to the percentage from the inclusions in felspar was given. In this paper the analyses of the total alpha particle activity per cm^2 of the major minerals (a) are compared to the radioactivity of the major minerals minus the activity contributed by the inclusions (b).

The ratios are given in Tables 1, 2, 3, 4, 5. An increase in the ratios indicates an increase of radioactivity from the inclusions. The ratio of a/b against the iron ratios (WAGER 1955) of a wide range of rocks is given in Fig. 5. The results represent 34 individual analyses. It is significant that the majority of the ratios lie between an a/b value of 1—3. The only values greater than 3 represent the acid and basic granophyres of Skaergaard, and the later G2 granites of Skye.

3) The distribution of uranium in the Skaergaard intrusion is given in Figs. 6, 7, together with the distribution of radioactivity in the minerals. The uranium content from the total felspar, and from inclusions in felspar is given in Fig. 6, and for the pyroxenes in Fig. 7. Uranium in the inclusions of felspar and pyroxene can be seen to increase towards

the late stage rocks. Uranium enrichment is also apparent in the perpendicular felspar rock E.G. 1851.

Some of the rocks at an altitude greater than 1,900 metres in the Skaergaard intrusion include the ones suspect of being weathered. In a previous paper (HAMILTON 1959) it was considered that a loss of c. 50 % uranium might account for the low uranium values in these

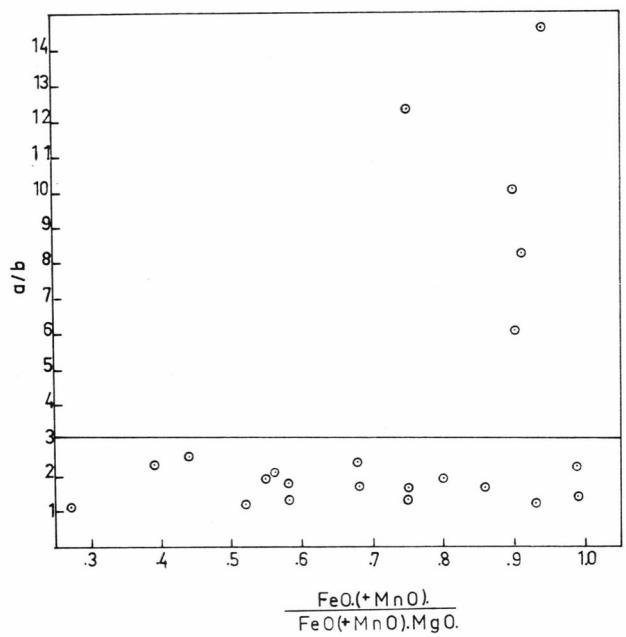


Fig. 5. The distribution of the iron ratios plotted against the value a/b.

rocks. The analyses of the major minerals make this supposition questionable. The uranium content of E.G. 4312 coincides with the appearance of primary apatite up to c. 6 % by volume. These apatites show an enrichment in uranium and thorium. The rock is quite fresh if compared with the weathered specimens E.G. 4330 and 3058. The enrichment of uranium in the apatite could lead to a depletion in the uranium content of the major minerals. The gradual increase of radioactivity in the later rocks would be interpretated as a later cyclic "build up". Although the primary minerals continued to grow at the expense of the intercrystal liquid it is not considered that the uranium and thorium enrichment in the apatites is due to the absorption of uranium and thorium from the intercrystal liquid.

4) The accessory minerals constitute the most radioactive minerals in the rocks studied.

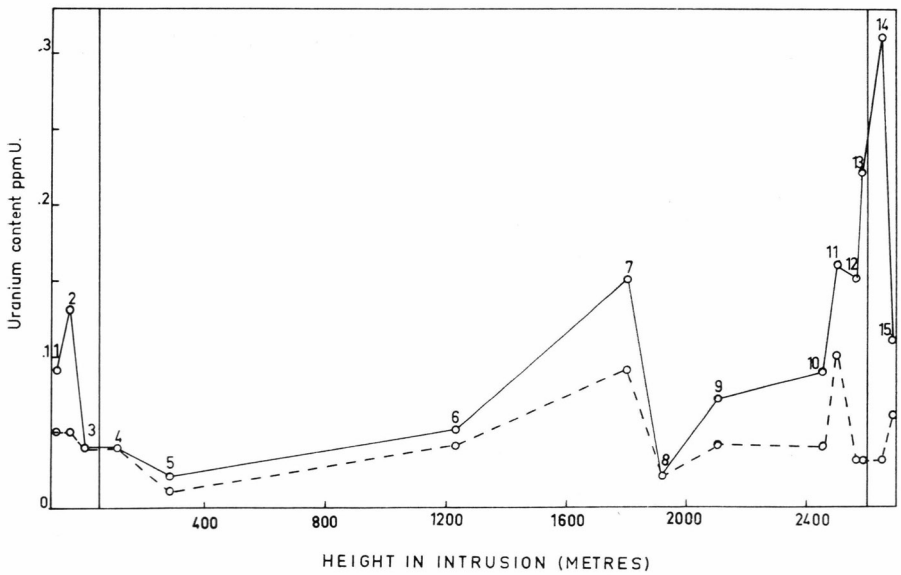


Fig. 6. Plot of the distribution of uranium in total felspar. (unbroken line) and in 'pure" felspar (broken line) against height in the Skærgaard intrusion.

Number sequence.

1. E.G. 4507 Chilled marginal gabbro.
2. E.G. 1851 Perpendicular felspar rock.
3. E.G. 4526 Gabbro picrite.
4. E.G. 5111 Hypersthene olivine gabbro.
5. E.G. 5086 Hypersthene olivine gabbro.
6. E.G. 4427 Middle Gabbro
7. E.G. 5181 Hortonolite ferrogabbro.
8. E.G. 4312 Ferrohortonolite ferrogabbro.
9. E.G. 4312 Ferrohortonolite ferrogabbro.
10. E.G. 4327 Fayalite ferrogabbro.
11. E.G. 4328 Fayalite ferrogabbro.
12. E.G. 4330 Basic hedenbergite granophyre.
13. E.G. 5259 Acid granophyre.
14. E.G. 5250 Transgressive granophyre.
15. E.G. 3058 Acid granophyre.

Rock numbers 1, 2, 3, 13, 14, 15 are not related to height in the intrusion.

5) Apatite is of interest as it is a constant accessory mineral in many rocks. McKeown (1956) showed that a direct by poor correlation exists between, P, U, Th, Rare Earths, and radioactivity. Monazite bastenaesite and haematite often fill fractures and form rims around some apatites. The distribution of uranium and thorium in fluor and chlor-apatites deserves attention. The concentration of uranium in organic phosphate material has been described by Bowie (1956) and Coppens (1956). Nuclear emulsion studies indicate that the most radioactive apatite

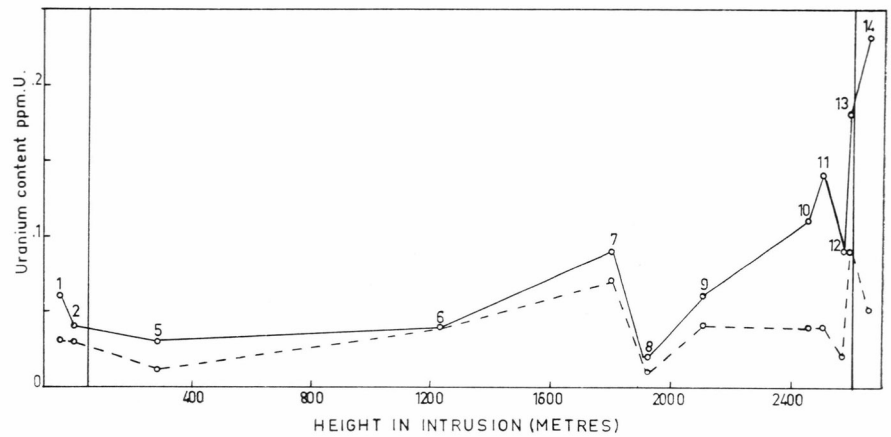


Fig. 7. Plot of the distribution of uranium in total pyroxene (unbroken line) and in "pure" pyroxene (broken line) against height in the Skærgaard intrusion. Number sequence as in Fig. 6.

generally occurs as very small crystals associated with iron ore, and not with the larger grains. The geochemistry of uranium in apatite and phosphorite is described by ALTSCHULER (1958).

6) The effect of "sampling" errors in the emulsion work is shown by the standard granite G1. The variations in the radioactivity of the thin section examined are correlated to the sporadic distribution of the accessory minerals. In more basic rocks such errors are far less. The analyses of several of the basic Skaergaard rocks (E.G. 4507, 5086, 4427) showed no significant variations. The main variations in basic rocks, in particular fine grained basalts, trachytes and dolerites can be related to variations in the amount of intercrystal activity. The extent to which ground water will affect the uranium content of a rock will vary considerably. Inadequate sampling, the irregular localisation of radio-elements, the effects of differentiation on the distribution of radio-elements (KEEVIL 1943) are all contributary causes. Radium enrichment in the uppermost 150 metres of a granodiorite laccolith has been reported by SOLOVIEV (1936).

Granulated acid igneous rocks have a higher alpha particle activity. Most of the activity can be removed with dilute acid (HURLEY 1950). It is probable that most of the leachable uranium decreases with depth out of range of the water saturation zone. The leaching studies of BROWN (1953) are of interest particularly the removal of 40 % of the total rare rarths. From preliminary observations orthite has proved to be an interesting mineral in its mode of occurrence. It has been observed that:

a) Dikes and veins cutting the Skaergaard intrusion (probably formed by remelting of country rock at depth) contain orthite.

b) The early G1 granophyres of Skye have orthite while the later G2 types have none. The G1 alpha particle activity is very similar to the acid differentiate of Skaergaard, while G2 is similar to "normal" acid igneous rocks, with an increase in radioactivity from inclusions in the felspar.

c) In Greenland (Ivigtut area) a zone of orthite-bearing rocks of the amphobilitic facies forms a band parallel to the foliation of the gneiss. (BERTELSEN 1959). If large amounts of the rare earths were present in intercrystal areas, and the rock were subjected to remelting, the first wave of material would be enriched in rare earths and orthite could possibly form.

The actual distribution of uranium and thorium atoms within a mineral is of particular interest. Unfortunately it is not possible to differentiate between uranium and thorium in a quantitative manner by the emulsion method. Kosov (1955) states that uranium and thorium occupy different positions in the lattice of a mineral in relation to the system of capilaries. The distribution of hydrated iron oxides, as films and as ferruginous hydrated iron oxides, is of importance in the geochemistry of uranium. Karhanavala (1958) describes metamict haematite. This shows that the uranium is present in the lattice of the haematite. On the other hand, magnetite though more susceptable to radiation damage, was non-metamict as the uranium was not present within the mineral lattice. The effect of iron oxide film is two-fold. In the final solidification of a rock uranium and thorium are adsorbed on to hydrated iron oxide films. In the decomposition of a rock or mineral, uranium and thorium are released. If decomposition is caused by hydrothermal solutions, the hydrated iron oxides will "adsorb" uranium and thorium Uranium so located is very susceptible to leaching by the action of ground water.

Preliminary determinations on the thorium content of the Skærgaard rocks indicates a U : Th ratio of 1 : 4. This only applies to the basic members.

APPENDIX

The distribution of radioactivity as shown by the nuclear emulsion method are given in the following diagrams and photographs.

Fig. 8.

1) A. A generalised section through a rock slice coated with nuclear emulsion. The emulsion is c. 40—50 microns thick, and the relation between the tracks formed at the surface of the emulsion to those formed from the rock slice are shown.

2) a. If the radioactivity originates from the surface of a crack in a mineral, or between minerals, the length of the tracks registered in the emulsion depend upon the thickness of mineral (absorbing medium) that the alpha particle has had to pass through.
b. Viewed through a microscope the tracks are longer at the junction of the crack to the emulsion. As the track dips away from the observer so the tracks registerd in the emulsion decrease in length. Finally the alpha particle emitted from the deeper parts of the crack are totally absorbed in the mineral and no track is formed in the emulsion.

3) a, b. Should the radioactive material constitute a point source a radial type of pattern of tracks is formed in the emulsion. The length of the track corresponds to the energy of the alpha particle. The "end points" of these tracks demarcate the familiar pleochroic radioactive haloes seen in biotites and other minerals.

4) a, b. A radial pattern of alpha tracks is often seen, but unlike the previous case the tracks do not converge to a point source. This is often caused by the prescence of a radiosource below the surface of the rock slice. By lowering the objective the projection of the tracks can often be seen to converge on an inclusion.

5) a. Shows a single alpha particle track originating from the surface of a mineral. By observing the end point of the track under a higher magnification a micro inclusion can often be seen (5 b).

6) a, b. The opaque minerals are often radioactive, by using a light source from below the alpha particles can only be seen if some of the tracks project beyond the edges of minerals. In cases of doubt the slides are re-examined using a vertical illuminator.

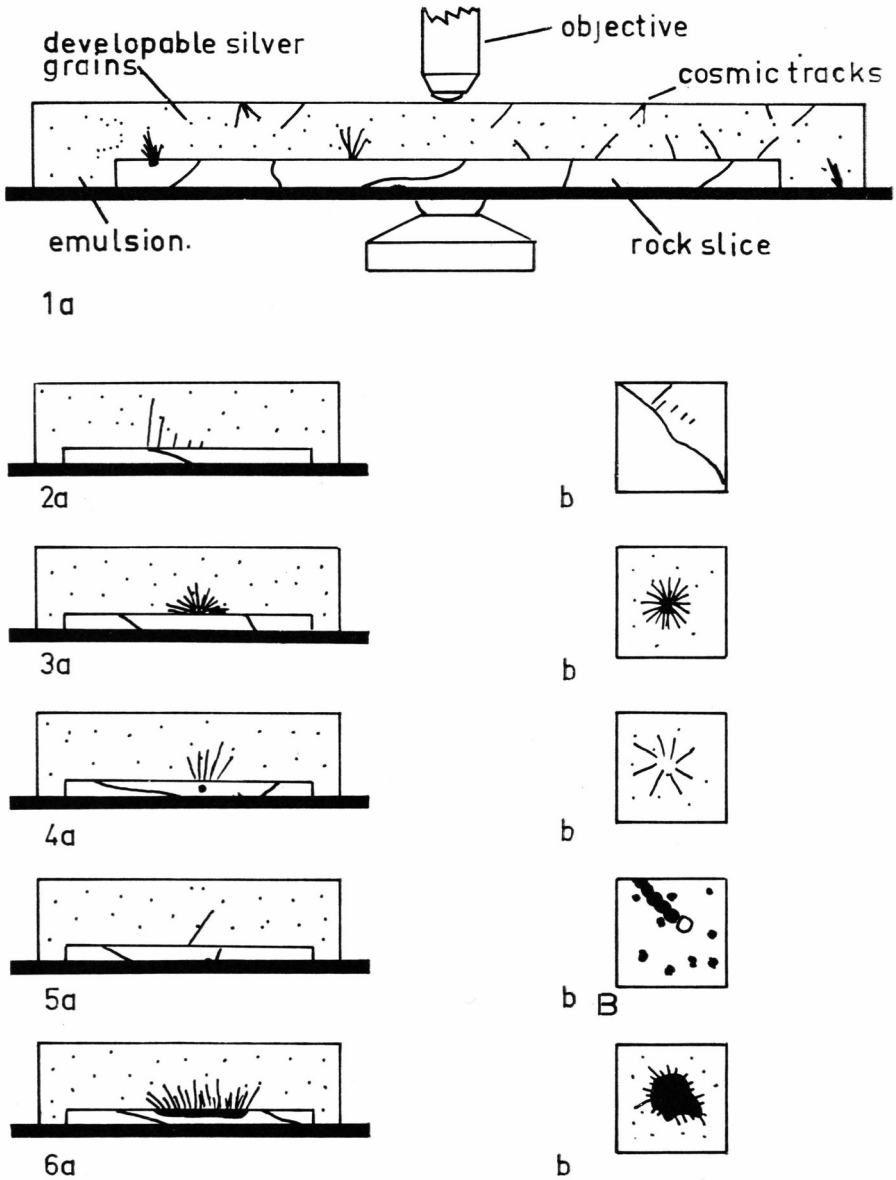

developable silver grains
objective
cosmic tracks
emulsion.
rock slice
1a
2a
b
3a
b
4a
b
5a
b
B
6a
b

Fig. 8.

Fig. 9—13.

Fig. 9. Zircon, Ivigitut Aplite. Greenland.

The distribution of radioactivity in zircon is extremely variable. In this case the central part of the crystals are highly radioactive, and are surrounded by a clean zone of zircon of low activity. The central parts are metamict and clouded by many inclusions of brown iron oxides. In the same rock purple fluorite is abundant but is almost inactive. The fluorite does contain radioactive inclusions surrounded by haloes.

Fig. 10. Zircon from G1 microgranites of Skye. Scotland.

Many of the zircons are zoned, but the radioactivity is too low to comment on the distribution of radioactivity. It is a feature of these rocks that some crystals are surrounded by a iron halo containing considerable amounts of radioactivity. It is possible that these zircons are infact xenotimes.

Fig. 11. Altered augite syenite Ilímaussaq, Greenland.

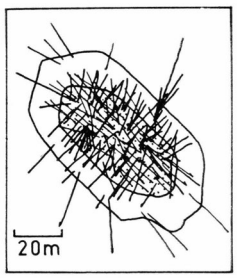
Uranium and thorium have been introduced into this rock by later hydrothermal solutions. The uranium and thorium is concentrated in intercrystal areas and with apatite. The radioactive mineral is so dusty as to make a mineralogical determination impossible.

Fig. 12. Altered augite syenite Ilímaussaq, Greenland.

Parts of the apatite in optical continuity with the large grains are highly radioactive. This only occurs in peripheral areas.

Fig. 13. Orthite, GI granophyre Skye.

The association of orthite and epidote is quite common in these rocks. The epidote is very weakly radioactive compared to the orthite.



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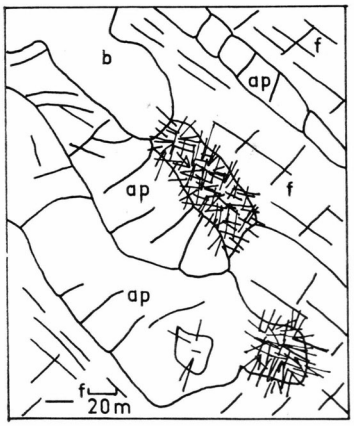

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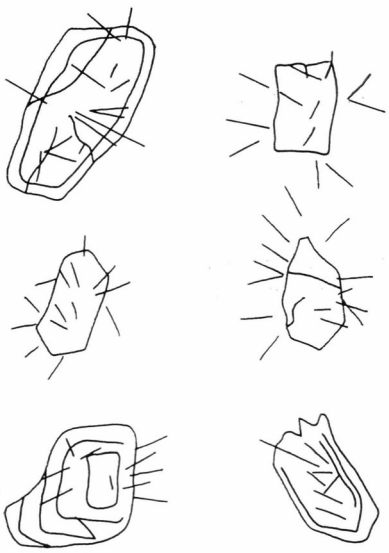

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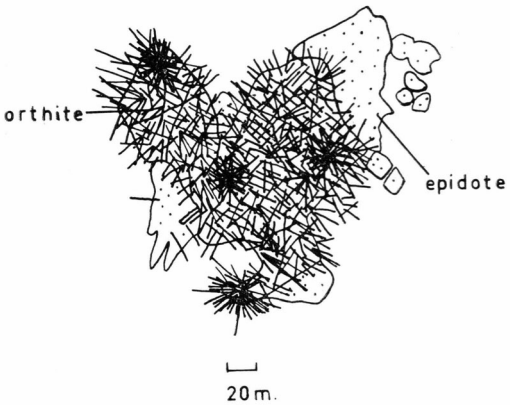

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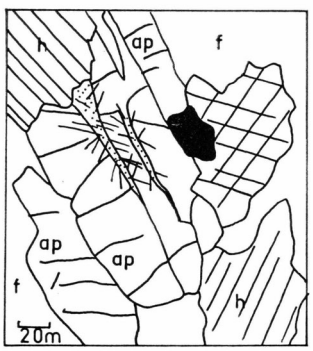

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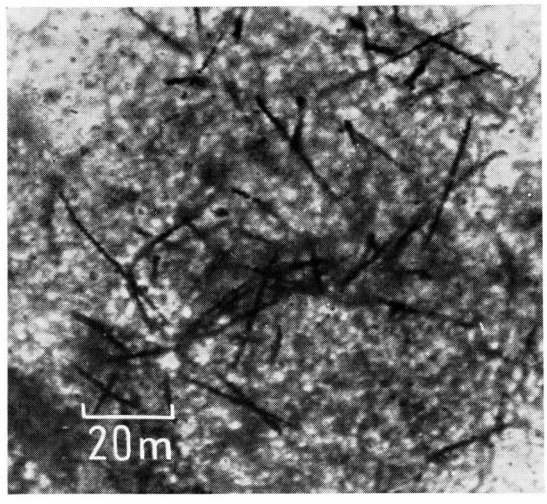
Fig. 9—13

Fig. 14—16.

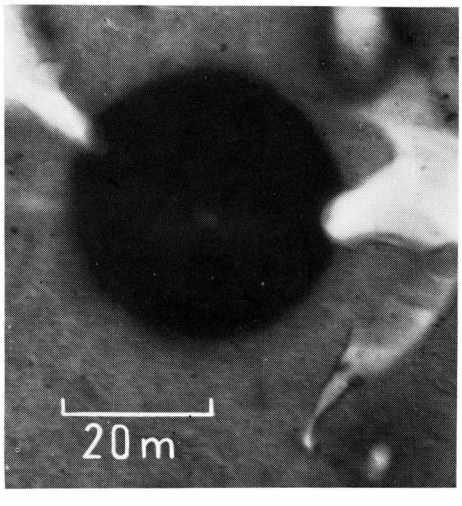
Fig. 14. Arfvedsonite granite Ilímaussaq, Greenland.
Alpha particle activity associated with ferruginous material.

Fig. 15. Julianehaab granite.
Pleochroic haloes in biotite.

Fig. 16. Julianehaab granite.
A radioactive inclusion in felspar. Radial cracks can be seen to surround the mineral. Although no pleochroic halo is present radial and circular zones surround the grain. These zones contain concentrations of ferruginous material. The radius of these zones corresponds to the haloes seen in biotite.



14



15

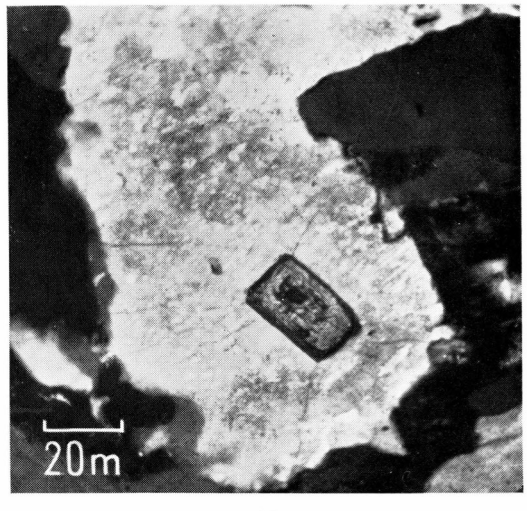


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Fig. 14—16.

Fig. 17—19.

Fig. 17. Lujavrite, Ilímaussaq, Greenland.

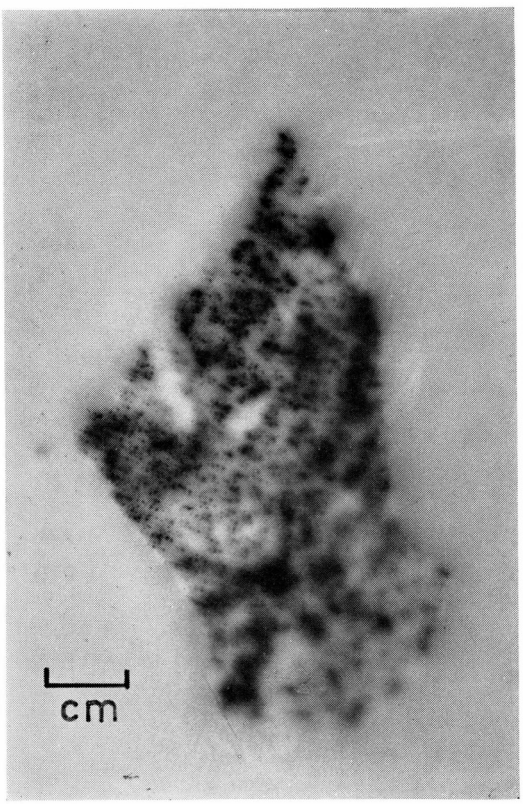
A cut specimen of lujavrite was placed in direct contact with a sheet of Kodak No-Screen X-Ray paper. The specimen was exposed for 18 days. The amount of blackening is approximately proportional to the total uranium and thorium content of the rock.

Fig. 18. Thorium and uranium mineralisation in a breccia, Agpat Ilímaussaq Greenland.

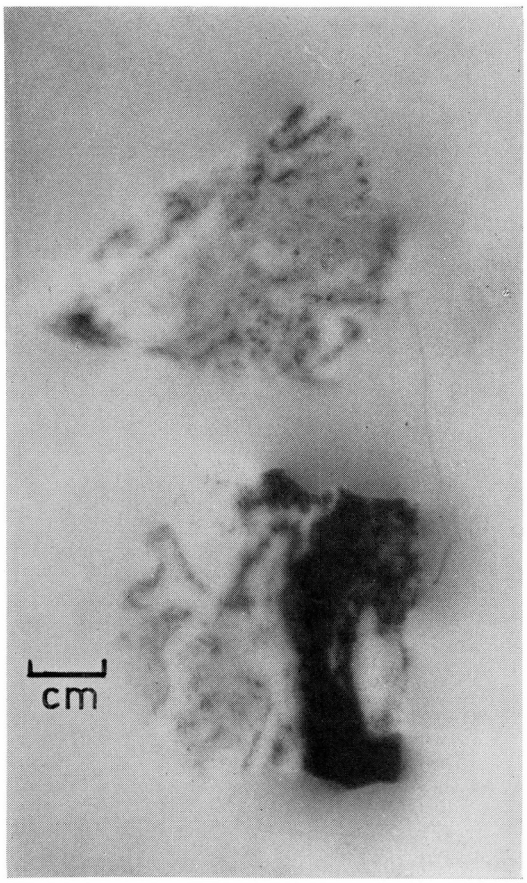
The method of exposure was similar to No. 15. This autoradiographic method has been of particular value in tracing the mineralisation in breccias, and in rocks showing small scale structural deformations.

Fig. 19. Altered felspar rock, Ilímaussaq intrusion Greenland.

The areas of blackening correspond to ferruginous areas containing fluorite.



17



18

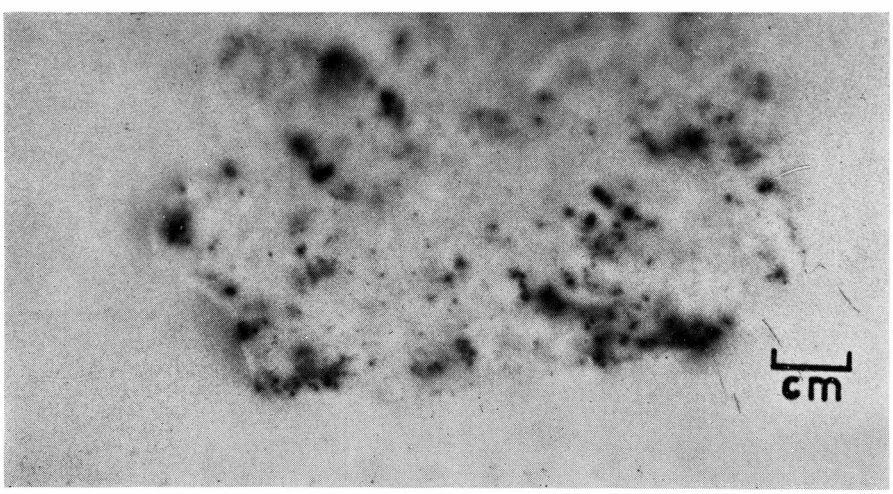


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Fig. 17—19.

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