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ABUNDANCE AND DISTRIBUTION OF URANIUM AND THORIUM IN ZIRCON, SPHENE, APATITE, EPIDOTE, AND MONAZITE IN GRANITIC ROCKS

By P. M. Hurley and H. W. Fairbairn

Trace Elements Investigations Report 636

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By Patrick M. Hurley and Harold W. Fairbairn*

ABSTRACT

Analyses were made of uranium and thorium in zircon, sphene, apatite, epidote, and monazite separated as accessory minerals from samples of granitic rock from widely scattered localities to indicate the abundance and distribution of these two elements among the five mineral phases. For any pair of mineral phases the distribution ratio remains within the same order of magnitude over the different rocks tested, although the variability of the data is such that only wide departures from constancy could be ascertained. Such gross differences have not been found. The approximate distribution of the two elements in the accessory mineral phases, taken in pairs, is as follows:

Minerals	Ratios Uranium	Ratios Thorium
Zircon/sphene	6.6	2.1
Sphene/apatite	4.7	2.9
Apatite/zircon	0.08	0.32
Monazite/apatite	> 23	> 530
Sphene/epidote	10	2.3
Zircon/epidote	34.3	3.0
Apatite/epidote	3.2	0.72

These data are mutually fairly consistent and the partition of the minor elements in the mineral phases remains roughly the same despite considerable variation in the absolute amounts of uranium and thorium present.

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INTRODUCTION

The principal host minerals for uranium and thorium in the earth's crust are zircon, sphene, apatite, epidote, and monazite, which occur as accessory minerals in crystalline rocks. In this investigation the object has been to obtain an approximate measure of the abundance and distribution of uranium and thorium among these five mineral phases.

Samples of the accessory minerals, generally about 99 percent pure, were obtained from a number of granitic rocks by standard gravity and magnetic methods (Fairbairn, 1955). The rocks are dominantly granite except as noted in the tables, and concentrates were obtained from samples of about 50 pounds. The uranium and thorium were measured by a method described by Hurley (1956) in which the 238 kev gamma ray from Pb^{212} in the thorium series is used to differentiate thorium against a total gamma-ray count in a two-channel scintillation spectrometer.

ERRORS

In the above reference also (Hurley, 1956), an estimate is given of the expectable errors in measurement. When the quantity of sample and activity is sufficient for a counting rate of at least twice background, the standard deviation error in measuring uranium or thorium is less than 10 percent if the constituent in question contributes an activity of at least 20 percent of the total. If the uranium activity is proportionately very low compared to the thorium, or vice versa, the error in its measurement becomes large. The uranium series has three times the activity of the thorium series, so that thorium values should be divided by three for a direct comparison of the relative activities of the two constituents.

For example, if the thorium content is 12 percent of the uranium content, its contribution to the total gamma count is only about 4 percent, and the estimated error in measurement of the thorium as given in the above-mentioned reference, is then 17 percent. On the other hand the error for the major constituent, uranium, is only 4 percent.

RESULTS

The results of the analyses are given in tables 1 to 7. Table 8 shows the average ratios of thorium to uranium in the different minerals. In table 9 examples are listed in which two minerals from the same rock have been analyzed, so that the distribution of uranium and thorium in the two phases can be observed.

In figure 1 the distribution of uranium and thorium in the accessory mineral phases is shown for three of these phases at a time. Only four examples are shown, but other combinations can be drawn from the average figures given in table 9. It was not possible to obtain three accessory phases from the same sample of granite so that the distribution of uranium or thorium in the different phases had to be obtained from samples yielding two of the phases at a time. Thus in figure 1A, for example, the distribution of uranium and thorium in zircon and sphene was obtained from the average of several granite samples as listed in table 9A. Similarly the distribution of uranium and thorium in zircon and apatite (table 9C), and in apatite and sphene (table 9B), was obtained independently from different sets of granite samples from rocks of differing ages in widely scattered localities.

Table 1.--Uranium and thorium content of accessory zircon from granitic rocks.1/

Sample no. 2/		Locality	Quarry	Uranium (ppm) 3/	Thorium (ppm) 3/
		Massachusetts			
3004	A	3 miles WSW of Peabody	Lineham	1210	375
	B	do.		510	205
	C	do.		405	130
3005	A	0.5 mile WNW of Rockport	Flat Ledge	2765	1890
	B	do.		2270	1200
	C	do.		1320	740
3006	A	2 miles WNW of Rockport	Blood Ledge	970	550
	B	do.		630	380
3011		2 miles W of Leominster	Leavitt	1130	380
3012	A	2 miles NE of Milford	Norcross	1350	480
	B	do.		955	445
3106	A	1.7 miles NNE of Milford	West	2400	915
	B	do.		2080	705
	C	do.		1425	490
3014	B	2.5 miles ESE of Wrentham		1325	680
3013	A	1.5 miles WNW of Uxbridge	Blanchard	2040	1090
	B	do.		1875	750
	C	do.		650	480
3052		8.2 miles N of North Attleboro		1820	900
3051		3 miles N of North Attleboro		1500	390
3107	A	North end of Whitinsville		1640	1000
3050	A	SW side of Hoppin Hill, North Attleboro		2000	2100
	B	do.		1540	1400
1982		Dedham, on Route 1		670	380
		Nova Scotia			
2090	A	Route 3 outside of Halifax		770	75
2093	A4/	Route 3, Birchtown, Shelburne County	Dauphine	1900	500
	B	do.		1550	450
2094		1 mile E of Atbood's Brook, Shelburne County, Route 3		680	60
2096	A	0.5 mile E of Albany Cross on Route 10, Annapolis County		830	160
	B	do.		620	75
2097		1 mile S of Springfield, Route 10		840	0
2098		3.5 miles N of New Ross, Route 12		850	100
2099	A	8 miles S of Kentville, Route 12, Kings County		630	100
	B	do.		560	180

Table 1.--Uranium and thorium content of accessory zircon from granitic rocks--Continued.

Sample no. 2/	Locality	Quarry	Uranium (ppm) 3/	Thorium (ppm) 3/
	Miscellaneous			
3086 A	Murray granite, Sudbury, Ontario		1700	460
B	do.		1200	280
3078	7 miles S of Calais on Route 1, Maine		2540	1100
3063	Hollingsworth and Whitney Road, 6 miles W of Route 201, Maine		1030	460
3087	Creighton granite, Sudbury, Ontario		3000	490
3125	Lausitz-massif, Germany		950	525
1819	Max Patch, N. C.		237	90
1644	Beach sand concentrate, N. C.		312	74
	Climax, Colo.5/		2450	1120

1/ All rocks are granites unless otherwise noted.

2/ Samples were separated into different fractions according to magnetic susceptibility, and these were designated A, B, and C in order of decreasing susceptibility.

3/ Errors in these analyses are discussed in the text.

4/ Quartz diorite.

5/ Silica-rich metasomatic rock.

Table 2.--Uranium and thorium content of coarse crystals of zircon from pegmatites.

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
1661	Iredell County, N. C.	580	500
1669	Buncombe County, N. C.	730	1650
1659	Renfrew County, Ontario	140	10
1660	Brudenell Township, Ontario	105	50
1795	Haliburton County, Ontario	3520	1080
1800	N. Hastings County, Ontario	3550	3020
1799	do.	4380	3840
1801	Madawaska, Ontario	25,200	1600
1805	Haliburton County, Ontario	5380	500
1797	Grattan Township, Ontario	6400	1050
1681	Leeds County, Ontario	81	93
1962	Wilberforce, Ontario (acid treated)	4000	(100)
1958	Oklahoma (nonmetamict)	300	675
1960	Oklahoma (metamict)	1550	7500
1678	Hammond, N. Y.	1570	600
1677	Lewis County, N. Y.	800	10
1915	Tigerville, N. C.	575	1020
1664	El Paso County, Colo.	480	2070
2088	South Africa	1020	355
1825	Litchfield, Maine	535	340
3038	Ceylon	5850	600

Table 3.--Uranium and thorium content of accessory sphene from granitic rocks.1/

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
3060	Mount Waldo, Maine	1270	165
3058	Sprucehead Island, Maine	105	20
3055	Hallowell, Maine	190	140
3062	5.5 miles S of Jackman, Maine, Route 201	120	430
3063	Hollingsworth and Whitney Road, 6 miles W of Route 201, Maine	250	375
3071	St. Gideon, Quebec	214	0
3079	1 mile S of Jonesboro, Route 1, Maine	39	80
3081	Barriefield, Ontario, near Kingston	165	600
3080	2 miles N of Mt. Desert, Route 198, Maine	245	950
3082 2/	6 miles N of Kingston, Ontario	70	150
3084	Gaspe, 7 miles S of Gaspesie, Quebec	55	150
3085	West end of Mountainville, Maine	300	1070
3051	3 miles N of North Attleboro, Mass., Route 1	160	90
3014	2.5 miles ESE of Wrentham, Mass.	155	325
3008	McCarthy quarry, Mass.	175	3150
2093 3/	Birchtown, Shelburne County, Nova Scotia, Route 3	280	180
3069 4/	St. Josef's Blvd., Montreal	25	90
1968 5/	Idaho	370	900
1969 3/	Idaho	206	190
1970 6/	Clayton Cone, Utah	400	560
	Alta Stock,6/ Utah	287	350
1971 7/	Blueberry Mountain, Mass.	1380	2430
1684 8/	Franklin, N. J.	215	190
1677 8/	Lewis County, N. Y.	218	170
1782 9/	Nipissing district, Ontario	127	70

1/ All samples are granites unless otherwise noted.
2/ Syenite.
3/ Quartz diorite.
4/ Tinguaite.
5/ Gneiss.
6/ No information.
7/ Metasomatic contact rock.
8/ Metamorphic rock.
9/ Pegmatite.

Table 4.--Uranium and thorium content of accessory apatite from granitic rocks.1/

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
2090	Route 3 outside of Halifax, Nova Scotia	95	80
2092	3.8 miles E of Port Monton, Nova Scotia	80	tr.
2093 2/	Birchtown, Shelburne County, Nova Scotia	30	30
2095	W. Nicteaux, Annapolis County, Nova Scotia	215	0
2096	0.5 mile E of Albany Cross, Route 10, Nova Scotia	90	80
2097	1 mile S of Springfield, Route 10, Nova Scotia	140	95
2098	3.5 miles N of New Ross, Route 12, Nova Scotia	105	80
2099	8 miles S of Kentville, Kings County, Nova Scotia	85	50
2100	Near Shelburne, Nova Scotia	85	400
3007	Harris quarry, Mass.	10	10
3008	McCarthy quarry, Mass.	150	5
3011	2 miles W of Leominster, Mass.	20	10
3042	Westerly, R. I.	10	20
3059	Lincolnville quarry, Maine	85	100
3055	Hallowell, Maine	55	80
3085	Mountainville, Maine	40	100
3056	N. Jay, Maine	50	50
3057	Waldoboro, Maine	50	10
3063	Hollingsworth and Whitney Road, Maine	60	120
3066	Scotstown, Quebec	160	5
3067	Brome Mountain, Quebec	25	85
3065	Mount Megantic, Quebec	50	75
3068 3/	Mount Johnston, Quebec	10	40
3070	Chicoutimi, Quebec, Route 54 S of Route 16A	30	60
3074	N side of Bagotville, Quebec	75	50
3078	7 miles S of Calais, Route 1, Maine	100	250
3073	Kenogami, Route 54, Quebec	tr.	tr.
3081	Barriefield, Ontario, near Kingston	10	20
3082 4/	6 miles N of Kingston, Ontario	5	5

1/ All samples are granites unless otherwise noted.

2/ Quartz diorite.

3/ Essexite.

4/ Syenite.

Table 5.--Uranium and thorium content of coarsely crystalline apatite.

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
1654	Durango, Mexico	10	250
1658	Wilberforce, Ontario	285	1600
1656	Bedford Township, Ontario	30	160
1657	Snarum, Norway	40	40
1655	Templeton, Quebec	20	260

Table 6.--Uranium and thorium content of monazite concentrates from granites (percentage concentration mostly 80 percent).

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
2092	3.8 miles E of Port Monton, Nova Scotia	3,000	39,000
2096	0.5 miles E of Albany Cross, Route 10, Nova Scotia	low	26,000
2100	Near Shelburne, Nova Scotia	low	75,000
3008	McCarthy quarry, Mass.	2,000	50,000
3042	Westerly, R. I.	low	75,000
3078	7 miles S of Calais, Route 1, Maine	1,200	33,500
3055	Hallowell, Maine	low	44,000
3056	N. Jay, Maine	2,500	51,000
3066	Scotstown, Quebec	2,500	50,000
3109	6 miles W of Lowell, Mass.	low	58,000
3107	Whitinsville, Mass.	1/ 50,000	45,000

1/ Probably a uranium-bearing mineral in concentrate.

Table 7.--Uranium and thorium content of accessory epidote from granites.

Sample no.	Locality	Uranium (ppm)	Thorium (ppm)
3063	Hollingsworth and Whitney Rd., Maine	27	160
3008	McCarthy quarry, Mass.	27	86
3062	Jackman, Maine	30	73
3051	3 miles N of N. Attleboro, Mass.	40	26
1982	Dedham, Mass.	43	185
3085	Mountainville, Maine	23	65
3058	Sprucehead Island, Maine	2	400
3014	Wrentham, Mass.	23	260
3079	Jonesboro, Maine	175	600

Table 8.--Average ratios of thorium to uranium in the different minerals.

Mineral	Ratio Th/U
Accessory zircon	0.4
Pegmatite zircon	1.0
Accessory apatite	1.3
Pegmatite apatite	10
Accessory sphene	1.7
Monazite	$\sim$ 25
Epidote	4.8

Table 9.--Distribution of uranium and thorium between pairs of accessory minerals from granitic rock samples.

Sample no.	Uranium (ppm)		Thorium (ppm)	
A.	Zircon	Sphene	Zircon	Sphene
3014	1325	155	680	325
3051	1500	160	390	90
2093	1700	280	475	180
3063	1030	250	460	375
Average	1390	210	500	240
Ratio, zircon to sphene	6.6		2.1	
B.	Sphene	Apatite	Sphene	Apatite
3058	105	40	20	60
3055	190	55	140	80
3063	250	60	375	120
3082	70	5	150	5
2093	280	30	180	30
Average	180	40	175	60
Ratio, sphene to apatite	4.7		2.9	
C.	Apatite	Zircon	Apatite	Zircon
3011	20	1130	10	380
2090	95	770	80	75
2093	30	1700	30	475
2096	90	720	80	120
2097	140	840	95	tr.
2098	105	850	80	100
2099	85	630	48	100
3063	60	1030	120	460
Average	80	960	70	215
Ratio, apatite to zircon	0.08		0.32	

Table 9.--Distribution of uranium and thorium between pairs of accessory minerals from granitic rock samples--Continued.

Sample no.	Uranium (ppm)		Thorium (ppm)	
D.	Monazite	Apatite	Monazite	Apatite
2092	3000	80	39,000	tr.
2096		90	26,000	80
2100		85	75,000	400
3008	2000	150	50,000	5
*3042		10	75,000	20
*3078	1200	100	34,000	250
*3055		55	44,000	80
*3056	2500	50	51,000	50
3066	2500	160	50,000	5
Average	2000	85	50,000	95
Ratio, monazite to apatite	> 23*		> 530*	

*Pure monazite values would exceed those for concentrate by several percent.

E.	Zircon	Epidote	Zircon	Epidote
3063	1030	27	460	160
3051	1500	40	390	26
1982	670	43	380	185
3014	1325	23	680	260
Average	1131	33	477	158
Ratio, zircon to epidote	34.3		3.0	

F.	Apatite	Epidote	Apatite	Epidote
3063	60	27	120	160
3008	150	27	5	86
3085	40	23	100	65
Average	83	26	75	104
Ratio, apatite to epidote	3.2		0.72	

Table 9.--Distribution of uranium and thorium between pairs of accessory minerals from granitic rock samples--Continued.

Sample no.	Uranium (ppm)		Thorium (ppm)	
G.	Sphene	Epidote	Sphene	Epidote
3063	250	27	375	160
3062	120	30	430	73
3051	160	40	90	26
3085	300	23	1070	65
3058	105	2	20	400
3014	155	23	325	260
Average	182	18	380	164
Ratio, sphene to epidote	10		2.3	

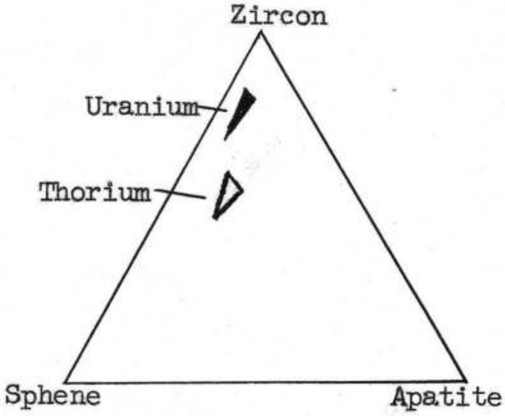

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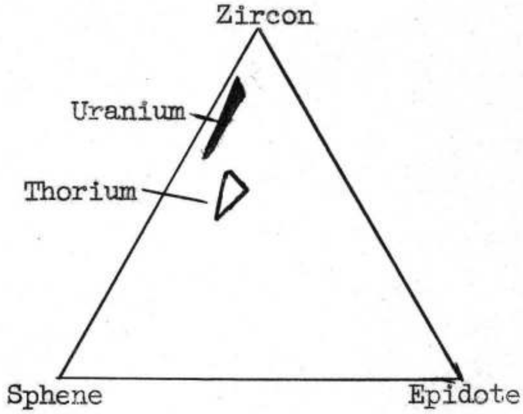
table 9A,B,C.



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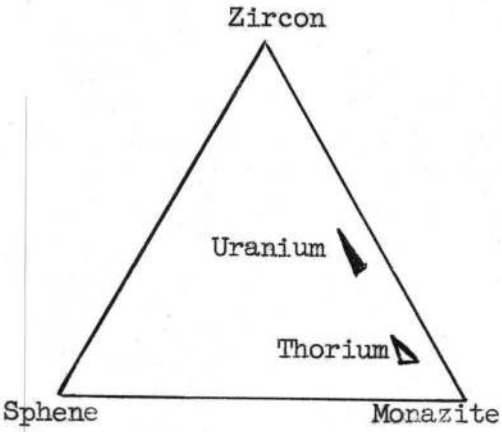

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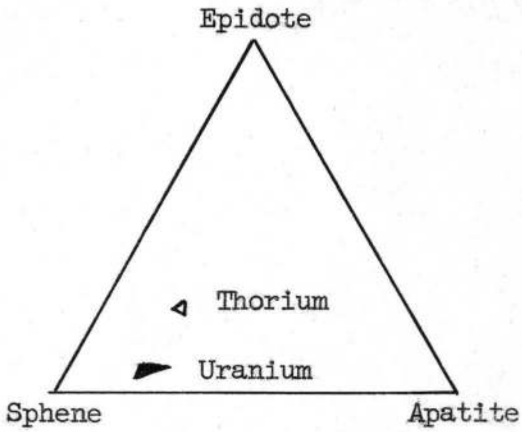

D. Data of
table 9B,F,G.

Figure 1.--Distribution of uranium and thorium among three accessory mineral phases for four combinations of accessory minerals.

In figure 1 the locations of uranium and thorium on the diagram, indicating their distribution in the accessory minerals, were obtained by the intersection of three lines drawn from each apex and dividing each base in the proportion found for the pair of minerals at either end of the base. The intersections of these lines form the triangles shown on the figure. The size of each triangle thus indicates how closely the lines came to intersection at a point, and thus is an indication of the analytical error or the constancy of the distribution of uranium and thorium in these three minerals. The values are averages only. Individual ratios may depart considerably from the average values, but it is difficult to tell at this stage of development of the analytical techniques how much is true variability and how much is due to precision errors in analysis. In monazite the values for the distribution have an uncertainty that results from a low proportion of inert impurities in the monazite concentrates and from the difficulty of measuring the uranium in the presence of such high proportions of thorium.

CONCLUSIONS

From these diagrams it is concluded that the partition of the uranium and thorium among the accessory mineral phases must have taken place under nearly equilibrium conditions, or else there would be less consistency in the results. Because the tolerance of the crystal structures of these five minerals for uranium and thorium (regardless of the mode of substitution) is undoubtedly a function of temperature and pressure, it cannot be expected that the distributions will be identical in each sample of granite. Furthermore, if the distribution factor giving the ratio of uranium in the solid phase to that in the fluid phase at time of crystallization is less than 1, the high concentration of uranium in the final interstitial fluid may upset the ratios because of the development of a late phase which may be intimately associated with the earlier rock-forming minerals. Evidence for this association is observed in autoradiographs of polished surfaces of granite which show concentration of uranium along grain boundaries, and also in acid leaching experiments (Hurley, 1950; Larsen and others, 1956) which demonstrate that this interstitial material containing the radioactive elements is soluble in dilute acids whereas most of the accessory minerals are not soluble.

Most granites have a ratio thorium/uranium of about 3 for the whole rock. Because zircon, sphene, and apatite have a smaller ratio than this, it seems likely that much of the interstitial radioactive material has a Th/U ratio higher than 3.

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