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NEW RESULTS FROM LEAD-ALPHA AGE MEASUREMENTS*

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ABSTRACT

Improvement in the spectrochemical method for determining lead in zircon increases the usefulness of the lead-alpha (Larsen) age method. Good agreement is found between the lead-alpha ages and those obtained by isotope dilution analyses on twelve samples. These samples have calculated ages ranging from 400 to 1200 million years. New lead analyses and revised lead-alpha ages are presented for 19 samples previously analyzed.

INTRODUCTION

A number of zircons previously analyzed by isotope Pb-U methods have been dated by the lead-alpha (Larsen) method using an improved spectrochemical procedure for determining lead (Rose and Stern, 1960). The new analytical technique differs from the previous method (Waring and Worthing, 1953) in that the standards used are more nearly similar in chemical and physical properties to natural zircons. Comparative results for lead determined by both spectrochemical methods indicate that the new analyses yield significantly higher lead values and lead-alpha ages than produced by earlier determinations.

ANALYTICAL DATA

The lead contents determined by spectrochemical and isotope dilution techniques and the measured and calculated alpha activities for 12 samples are given in Table 1. The table lists all Precambrian samples presently available and presents comparative results for splits of the same sample.

The average deviation between the spectrochemical lead values and the lead contents determined by isotope dilution analyses is about 2% with the isotope dilution values on the average greater. The average deviation between measured and calculated alpha activity is about 6% with the calculated activities generally higher.

A sample of monazite, SQ-81, from Mountain Pass, San Bernardino County, California, which had been previously dated by isotope dilution analysis and by the lead-alpha method was investigated in the present study. A lead content of 1760 ppm was obtained with the new spectrochemical method compared with an average value of 1130 ppm found previously (Jaffe, 1955, p. 1253). The present determination agrees with isotope dilution analyses by G. R. Tilton and L. R. Stieff who found 1770 and 1740 ppm of lead, respectively (Gottfried and others, 1959, p. 25).

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TABLE 1.—COMPARISON OF LEAD CONTENTS AND ALPHA ACTIVITIES

Sample No.	Zircon sample, and supplier	Pb(ppm)		Alpha activity		Th:U
		Spectro-chemical method[1]	Isotope dilution	Measured	Calculated from isotope dilution U and Th[2]	
1	Kensington granite gneiss, Washington, D. C., G. R. Tilton (Cooke, C. W., 1951).	97[3]	98	568	567	.20
2	Rare-earth vein, Laurel Gap, Tenn., G. R. Tilton	165	161	697	684	.31
3	Beech granite, Roan Mountain, Tenn., G. R. Tilton	54	58	235	242	.67
4	Granite gneiss, Crossnore, N. C., G. R. Tilton	33	32	100	99	.14
5	Gneiss, Shenandoah, Park, Va., G. R. Tilton	78	83	183	179	.32
6	Cranberry gneiss, Deyton Bend, N. C., G. R. Tilton	57	60	124	129	.43
7	Storm King granite, N. Y., G. R. Tilton (Berkey, C. P., 1907)	335	337	676	838	.16
8	McDonald mine Hybla, Ontario, G. R. Tilton	970[4]	1045	1488	1743	.59
9	Baltimore gneiss, Spring Mills, Pa., G. R. Tilton	187	168	372	390	.32
10	Pegmatite, San Gabriel Mts., Calif., L. T. Silver	37	41	78	81	.30
11	Old Whitestone Farm, Natural Bridge, N. Y., A. F. Buddington and H. D. Holland	110[5]	127	224[6]	283	—[7]
12	Wilson Creek gneiss, Mortimer, N. C., Tilton	168	160	433	484	.26

[1] Average of duplicate determinations of a 15 mg. sample.

[2] Sample from San Gabriel Mts., California isotopically analyzed by L. T. Silver, California Institute of Technology. All other samples isotopically analyzed by G. R. Tilton and G. L. Davis, Geophysical Laboratory, Carnegie Institute of Washington. Alpha activities were calculated from the analyses.

[3] Analyses by Nola B. Sheffey, U. S. Geological Survey, Washington, D. C.

[4] Sample diluted with 8 parts zircon base (68 per cent zirconia and 32 per cent silica) prior to lead analysis.

[5] Single determination on a 4 mg sample.

[6] Alpha activity measurement by H. W. Jaffe, U. S. Geological Survey, Washington, D. C.

[7] All alpha activity due to uranium, thorium not detectable.

In addition to the 12 samples for which isotope dilution analyses were available, new lead determinations were made on some samples previously dated by the lead-alpha method for which sufficient material was available (Table 3).

DISCUSSION

The new lead-alpha ages (Table 2) are in good agreement with the ages obtained by isotopic methods. The results for the 19 samples in Table 3 indicate, however, that although some of the lead analyses previously

TABLE 2.—COMPARISON OF LEAD-ALPHA AGES OF ZIRCON WITH LEAD-URANIUM AND LEAD-THORIUM AGES IN MILLIONS OF YEARS

Sample No.	Sample	Pb-Alpha: Assumed Th:U ratio 1:1	Pb-Alpha: Calculated Th:U ratio	$\frac{Pb^{206}}{U^{238}}$	$\frac{Pb^{207}}{U^{235}}$	$\frac{Pb^{207}}{Pb^{206}}$	$\frac{Pb^{208}}{Th^{232}}$	Reference
1	Kensington granite gneiss, Washington, D. C. (Cooke, 1951)	410	430	400	420	510	350	G. R. Tilton and others (1959)
2	Rare-earth vein, Laurel Gap, Tennessee	560	580	585	640	820	360	"
3	Beech granite, Roan Mountain, Tennessee	540	560	555	585	700	425	"
4	Granite gneiss, Crossnore, North Carolina	770	790	690	720	800	680	"
5	Gneiss, Shenandoah National Park, Virginia	980	1000	1070	1100	1150	1110	"
6	Cranberry gneiss, Deyton Bend, North Carolina	1040	1070	1080	1140	1270	950	"
7	Storm King granite, Bear Mt. Near York. (Berkey) 1907)	1120	1140	960	990	1060	850	G. R. Tilton and others (1958)
8	McDonald mine, Hybla Ontario	1420	1430	1350	1190	900	435	G. L. Davis and others (1957)
9	Baltimore gneiss. Spring Mills, Pennsylvania	1130	1160	1010	1045	1120	950	G. R. Tilton, written communication
10	Pegmatite, San Gabriel Mts., California	1070	1100	1200	1200	1200	1210	L. T. Silver and others (1960)
11	Old Whitestone Farm Natural Bridge, N. Y.	1100	1130	1025	1065	1140	—	G. R. Tilton and others (1957)
12	Wilson Creek gneiss, Mortimer, North Carolina	890	920	800	860	1020	670	G. R. Tilton and others (1959)

reported are in satisfactory agreement with the new determinations, most of the new determinations are significantly higher than the earlier lead analyses. Thus the new lead-alpha ages for these samples are significantly older. No systematic variation has been found between the old and new determinations and hence no single empirical factor can be applied to the earlier analyses.

The new lead-alpha ages are reported to the nearest 10 million years. The analytical error is assessed at approximately 10 per cent. This error is assigned to deviations in the spectrochemical and counting techniques and excludes errors due to variations in Th/U and geological factors that are more difficult to evaluate. The lead-bearing minerals other than zircon, present as a sample contaminant, are generally eliminated during careful microscopic examination prior to analysis. A more difficult problem is that of nonradiogenic lead contained within the zircon sample

TABLE 3.—REDETERMINATION OF SOME LEAD-ALPHA AGES

Rock types, locality and supplier	α/mg/hr.	Mean lead ppm (duplicate determinations)		Lead-alpha ages Million years		Remarks
		Previous method	Present method	Previous method	Present method	
Nordmarkite N-7, Oslo, Norway, Henry Faul	177[1]	19		265		C. L. Waring, oral communication (1960)
	173		18		260	
Reddish biotite granite SA-1, Jebal Rafa, Saudi Arabia, G. F. Brown	3,840	390	560[2]	247	350	Jaffe, and others (1959)
Coarse gray porphyritic granite, SA-2, Eastern side of eastern batholith, Saudi Arabia, G. F. Brown	460	66.5	117[2]	349	600	Jaffe, and others (1959)
	360	48	125[2]	323	800	
Granite rock, SA-3, Jebal Zaba, Saudi Arabia, G. F. Brown	2,083	303	530[2]	351	600	Jaffe and others (1959)
	1,970	307	570[2]	377	680	
Swarthmore granodiorite, A.W.P.-5, Intermediate replacement type East Lake Park, Philadelphia area, Pa. A. W. Postel	220	21.5	44[3]	238	480	Jaffe and others (1959)
Swarthmore granodiorite A.W.P.-6, Intermediate replacement type, Clifton Heights, Philadelphia area, Pa., A. W. Postel	250	23.5	48[3]	230	460	Jaffe, and others (1959)
Biotitic Wissahickon schist, A.W.P.-7, near Falls Bridge, Fairmont Park, Philadelphia area, Pa., A. W. Postel	135	23.5	52[3]	418	880	Jaffe, and others (1959)
Biotitic Wissahickon schist, A.W.P.-10, Gully Run, South of West Manayunk, Philadelphia area, Pa., A. W. Postel	125	22	45[3]	422	840	Jaffe, and others (1959)
Arenite, RN-2, Ocoee series, Great Smoky Mts., Gatlinburg quadrangle, Tennessee, R. B. Neuman	287	107	112[3]	850	890	Jaffe, and others (1959)
Arenite, RN-13, Ocoee series, Great Smoky Mts., Tunderhead quadrangle, Tennessee, North Carolina, R. B. Neuman, D. Carroll	136	37	48	640	820	Jaffe, and others (1959)
Quartz diorite, SV-1, Roadcut, north edge of town of San Vicente, Baja, California, D. Gottfried, L. R. Stieff, and T. W. Stern	123	5.05		102		Jaffe, and others (1959)
	152		6.1		100	
Baltimore gneiss, BL-1, Cross-cutting pegmatite along a deformed fault plane, River Road, Southeast of Spring Mill, Pa., Betsy Levin	160	44		654		D. Gottfried, personal communication
	141		52		850	

(*continued on next page*)

[1] Alpha activity measured by H. W. Jaffe.
[2] Spectrographic examination of 5 mg sample, single determination.
[3] Spectrographic examination on 15 mg sample, single determination.

TABLE 3 (*continued*)

Rock types, locality and supplier	α/mg/hr.	Mean lead ppm (duplicate determinations)		Lead-alpha ages Million years		Remarks
		Previous method	Present method	Previous method	Present method	
Baltimore gneiss, BL-2. Pegmatized band, River Road, Southeast of Spring Mill, Pa., Betsy Levin	144 130	41	61	670	1060	D. Gottfried, oral communication (1960)
Baltimore gneiss, BL-3, Concordant felsic band, River Road, southeast of Spring Mill, Pa., Betsy Levin.	191 173	51	81	630	1060	D. Gottfried, oral communication (1960)
Baltimore gneiss, BL-4, Light felsic band in gneiss, 1 mile southeast of Spring Mill, Pa., Betsy Levin.	237 228	62.5	105	620	1040	D. Gottfried, oral communication (1960)
Baltimore gneiss? BL-5, Concordant felsic garnetiferous band, north side of Glen Mills Quarry, Glen Mills, Pa., Betsy Levin.	663 608	104	215	380	820	D. Gottfried, oral communication (1960)
Baltimore gneiss? BL-6, Cross-cutting folded pegmatite in altered gabbro. Glen Mills quarry, Glen Mills, Pa., Betsy Levin.	234 211	41	44	405	500	D. Gottfried, oral communication (1960)
Cranberry gneiss, Deyton Bend, North Carolina, G. R. Tilton	124	39	57	734	1040	D. Gottfried, oral communication (1960), Tilton and others (1959)
Scarn at Old Whitestone Farm, Natural Bridge, New York, A. T. Buddington, H. D. Holland	224	66	110	771	1100	G. R. Tilton and others, (1957)

(Tilton and others, 1957). In addition, igneous rocks may contain mixed zircons, and the possible effects of the xenocrysts on the lead-alpha age are not easily evaluated. Any of these factors would tend to give older lead-alpha ages.

The calculated alpha activities in Table 1 were derived from the equation

$$\alpha = 0.366\,\mathrm{U} + 0.089\,\mathrm{Th}$$

where α is in units of alpha counts per milligram per hour, and the Th and U contents are in parts per million.

The age equations (Gottfried and others, 1959, p. 14–17) are

$$t = \left[\frac{2632 + 624\,\mathrm{Th:U}}{1 + 0.312\,\mathrm{Th:U}}\right]\frac{\mathrm{Pb}}{\alpha} = C\,\frac{\mathrm{Pb}}{\alpha}$$

and

$$t_0 = t - 1/2\, kt^2$$

The age equation, t, is used for samples which are younger than 200 million years and, t_0, for samples which are older than 200 million years. For most zircons, the Th:U ratios vary within an order of magnitude of a 1:1 ratio, and in the absence of thorium and uranium analyses, ages are calculated based on a 1:1 Th:U ratio. This assumption gives a value of 2485 for C and introduces an error in the calculation when the Th:U ratio deviates from 1:1. When the Th:U ratio is actually less than 1:1, the apparent age will be younger than the age derived from the actual ratio. This situation exists for the 12 samples given in Table 2. Both of the calculated lead-alpha ages are reported. The actual Th:U ratios are given in Table 1. The maximum error introduced by assuming a fixed 1:1 ratio is about 5 per cent for the samples reported. Recent work at the U. S. Geological Survey has shown that Th:U ratios may be measured by x-ray fluorescence to a lower limit of 50 ppm for each element. The Th:U ratios may then be used in the age calculation.

Based on the assumption that lead in zircon is primarily of radiogenic origin, the lead-alpha method has been considered to be most nearly comparable to the Pb^{206}/U^{238} isotopic age. As indicated in Table 2, when the Pb-U ages are concordant, the lead-alpha age may be expected to agree favorably. It is known, however, that the Pb/U ages obtained from zircon concentrates of Precambrian rocks commonly are discordant. Few data are at present available for discordant zircons and the correlation between lead-alpha and discordant Pb-U isotopic ages remains to be investigated.

The lead-alpha method is of great value as a reconnaissance tool. Its speed and simplicity are particularly advantageous. Only small amounts of zircon are required; 75 milligrams for the nondestructive alpha measurement, and 15 milligrams for each lead determination. With the refinement recently achieved in the determination of lead in zircon, the usefulness of the technique will be extended further. The method is of particular value for preliminary scanning of zircon samples prior to isotopic analysis and is a useful supplement to potassium-argon and rubidium-strontium age determinations.

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