INFORMATION TO USERS

This manuscript has been reproduced from the microfilm master. UMI films the text directly from the original or copy submitted. Thus, some thesis and dissertation copies are in typewriter face, while others may be from any type of computer printer.

The quality of this reproduction is dependent upon the quality of the copy submitted. Broken or indistinct print, colored or poor quality illustrations and photographs, print bleedthrough, substandard margins, and improper alignment can adversely affect reproduction.

In the unlikely event that the author did not send UMI a complete manuscript and there are missing pages, these will be noted. Also, if unauthorized copyright material had to be removed, a note will indicate the deletion.

Oversize materials (e.g., maps, drawings, charts) are reproduced by sectioning the original, beginning at the upper left-hand corner and continuing from left to right in equal sections with small overlaps.

**ProQuest Information and Learning
300 North Zeeb Road, Ann Arbor, MI 48106-1346 USA
800-521-0600**

UMI[®]

THE RICE INSTITUTE

THE RELATIONSHIP BETWEEN THE PETROLOGY AND
THE THORIUM AND URANIUM CONTENTS
OF SOME GRANITIC ROCKS

by

John M. Whitfield

A THESIS
SUBMITTED TO THE FACULTY
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

Houston, Texas
April 1958

*approved by
John J. W. Rogers*

UMI Number: 3079926

UMI[®]

UMI Microform 3079926

**Copyright 2003 by ProQuest Information and Learning Company.
All rights reserved. This microform edition is protected against
unauthorized copying under Title 17, United States Code.**

**ProQuest Information and Learning Company
300 North Zeeb Road
P.O. Box 1346
Ann Arbor, MI 48106-1346**

"Granite to the geologist is classic ground: from its widespread limits, and its beautiful and compact texture, few rocks have been more anciently recognized. Granite has given rise, perhaps, to more discussion concerning its origin than any other formation. We generally see it constituting the fundamental rock, and, however formed, we know it is the deepest layer in the crust of this globe to which man has penetrated. The limit of man's knowledge in any subject possesses a high interest which is perhaps increased by its close neighbourhood to the realms of imagination."

— Charles Darwin, 1837

ACKNOWLEDGMENTS

This research is a part of a larger investigation of the geochemistry of thorium, made possible through grant C-009 from the Robert A. Welch Foundation. I wish to express my gratitude to many individuals who aided materially in this study. The investigation was carried out under the direction of Dr. John J. W. Rogers, whose advice on all phases of the work was greatly appreciated. Dr. John A. S. Adams gave valuable suggestions and aid on all parts of the work, particularly on the analytical methods. Mr. William Orr and Dr. Jasper E. Richardson, of the Shell Development Company, gave extremely helpful advice on the design and calibration of the gamma-ray spectrometer. Mr. Richard Pliler kindly analysed many of the granites and provisional sedimentary standards by chemical methods. Dr. S. S. Goldich supplied several samples of granite from northern Minnesota and Canada. Thomas Barbee acted as field assistant during the collection of the remainder of the samples.

INTRODUCTION

The object of this investigation is to find large scale variations in thorium and uranium contents of granitic rocks and to link these variations to petrographic types and to geological characteristics of the plutons in which the rocks occur; in short, the object is to find the relation between the major petrogenesis of granitic rocks and their thorium and uranium contents. For this purpose representative granitic plutons from North America have been sampled. These samples have been examined petrographically and have been analyzed by gamma-ray spectrometry for thorium, uranium, and potassium. The study is predicated on the assumption that samples sufficiently representative of plutons could be obtained so that variation between hand specimens would not mask larger variations. The fact that large-scale relations are found justifies this assumption, at least in part.

The material will be presented in four main parts: a review of the previous literature, the analytical method, presentation of data, and results.

CONTENTS

	page
PREVIOUS WORK	2
OBJECTS AND METHODS	2
PRESENTATION OF DATA	4
I Sampling Homogeneity	9
<u>Sites of activity:</u>	9
<u>Empirical variation:</u>	12
II Distribution Within Plutons	15
III Distribution Within Comagmatic Series	21
IV Distribution in Rock Types Above the level of Comagmatic Series	22
CORRELATION	34
ANALYTICAL METHOD: GAMMA-RAY SCINTILLATION	40
GENERAL THEORY	40
I Nature and Origin of Gamma-Rays.	40
II Interaction of Gamma-Rays with Matter	43
<u>Photoelectric absorption:</u>	43
<u>The Compton effect:</u>	44
<u>Pair formation:</u>	45
III Natural Gamma-Ray Emitters	49
IV Statement of Principle	54
INSTRUMENTATION	55
I Detector	55
II Spectrometer	58

	page
OPERATION	52
I Calibration	52
II Spectra of the Natural Series	54
III Choice of Counting Channels	56
IV Width of Operational Window	72
V Drift Correction	74
VI Choice of Geometry and Standards	75
VII Calculations	80
VIII Preparation of Samples	85
IX Error, Accuracy, and Length of Count	87
<u>Precision:</u>	87
<u>Accuracy:</u>	92
COMPARISON WITH OTHER GAMMA-RAY SPECTROMETERS	94
PRESENTATION OF DATA	96
RESULTS	107
ANALYSIS OF DATA	107
DISCUSSION	118
CONCLUSIONS	121
APPENDIX	141
BIBLIOGRAPHY	160

ILLUSTRATIONS

Figure		page
1	Energy level diagram for Ra^{224}	42
2	Gamma scintillation spectrum for potassium. (From Hurley 1956)	50
3	Decay scheme of K^{40} to A^{40} and Ca^{40} .	54
4	Gamma-ray detector and shield	57
5	Calibration curves for the gamma-ray spectrometer	65
6	Gamma-ray scintillation spectrum for pure uranium series in secular radioactive equilibrium	67
7	Gamma-ray scintillation spectrum for thorium series in secular radioactive equilibrium	68
8	Gamma-ray scintillation spectrum for K_2CO_3	69
9	Compound spectrum: Uranium, thorium, and potassium spectra superimposed. .	70
10	Gamma scintillation spectrum for thorium series in equilibrium. (From Hurley, 1956)	71
11	Uranium content vs. thorium content of granitic rocks analysed in this investigation. (See table 14)	123
12	Thorium content vs. Th/U ratio of granitic rocks analysed in this investigation. (See table 14)	124
13	Uranium content vs. Th/U ratio of granitic rocks analysed in this investigation. (See table 14)	124
14	Uranium content vs. Th/U ratio of granitic rocks reported in the literature. (See tables 6 and 8)	125

Figure		page
15	Thorium content vs. Th/U ratio of granitic rocks reported in the literature. (See tables 6 and 8) .	126
16	Uranium content vs. thorium content of granitic rocks from the literature superimposed on those analysed in this investigation	127
17	Uranium content vs. Th/U ratio of granites reported in the literature superimposed on those analysed in this investigation	128
18	Thorium content vs. Th/U ratio of granitic rocks reported in the literature superimposed on those analysed in this investigation. . .	129
19	Uranium content vs. potassium content of granitic rocks analysed in this investigation	130
20	Thorium content vs. potassium content of granitic rocks analysed in this investigation	130
21	Potassium content vs. Th/U ratio of granitic rocks analysed in this investigation	131
22	Uranium and thorium contents vs. potash feldspar/plagioclase ratio (R) of granitic rocks studied	132
23	Th/U ratio vs. R of granitic rocks studied	133
24	Uranium content vs. color index of granitic rocks studied	134
25	Thorium content vs. color index of granitic rocks studied	134
26	Th/U ratio vs. color index of granitic rocks studied	135

Figure		page
27	Uranium content vs. quartz content of granitic rocks studied	136
28	Thorium content vs. quartz content of granitic rocks studied	136
29	Thorium content vs. anorthite content of plagioclase in the granitic rocks studied	137
30	Uranium content vs. anorthite content of plagioclase in the granitic rocks studied	137
31	Radiometric and petrographic para- meters plotted against R	138
32	Uranium and thorium contents of a group of intermediate and granitic rocks from several geographic regions plotted against Th/U ratio. (From Senftle and Keevil, 1947)	139
33	Schematic representation of postu- lated loss of uranium relative to thorium during normal differentiation	140

TABLES

Table		page
1	Th/U values for individual rocks. Arranged in order of increasing Th/U ratio	5
2	Distribution of uranium and alpha activity within individual plutons . .	16
3	Distribution of uranium and alpha activity in members of three magma series of the western U.S.	24
4	Distribution of uranium and alpha activity in members of four New Hampshire magma series	25
5	Distribution of alpha activity in North American rock types	28
6	Estimates of large areal activities, including world averages	30
7	Uranium and thorium in Russian granites	31
8	Thorium and uranium contents and alpha activity of rock types by geographic region (after Senftle and Keevil, 1947) . . .	32
9	Per cent standard deviation for Senftle and Keevil's regional variation data .	33
10	Estimates of world Th/U ratio averages	35
11	Natural radioactive isotopes	49
12	Comparison of estimated relative analytical error (precision) calculated by using statistical considerations compared with observed reproducibility	91
13	Interlaboratory and intralaboratory reproducibility of uranium, thorium, and potassium determinations	93.5
14	Thorium and uranium contents of all granitic rocks analysed in this investiga- tion, arranged in order of increasing Th/U ratio	97

Table		page
15	Modal analyses of granitic rocks, arranged in decreasing order of R . .	99
16	Comparison of radioactive and petro- graphic averages for various groupings of samples	116

PREVIOUS WORK

Analysis of published data is undertaken for two major purposes; the first is to collect data which have direct bearing on the problem, i.e. uranium and thorium contents and ratios and particularly any correlation of these data with petrographic type or geological characteristics of the rocks.

The second major purpose of analyzing the literature is to test, in-so-far as possible, the assumption upon which this investigation is based: that representative samples may be collected. If the unit of sampling necessary for reproducibility within petrographically identical rocks is larger than approximately 1000 grams, or if there are within-pluton variations in radioactivity or uranium and thorium contents which are not related to petrographic variations, then the diffuseness of the relationships on a larger scale may be attributed in part to the small-scale variation and not entirely to imperfection in major petrochemical control.

OBJECTS AND METHODS

Radioactivity and uranium and thorium data have been collected in the past for various reasons and by different methods. Most of the earlier work was done for three major purposes: 1) Calculation of heat produced in crustal rocks

(Poole and Joly, Joly, Piggot, Jeffreys and others),
2) thorium and uranium contents and their relation to the isotopic compositions and origin of lead (see papers by Holmes and Keevil), and 3) thorium and uranium contents and their relation to helium age determination (see papers by Keevil). Uranium and thorium only recently have been of geochemical and petrologic interest.

Most radioactivity data have been determined by radiometric techniques. Radium measurement by ionization due to radon gas (Poole and Joly, 1924; Piggot, 1931; and others) and the determination of total activity by alpha counting (Finney and Evans, 1935; and many others) are relatively simple, analytically; and the bulk of the available data has been obtained by these methods. Thoron streaming (Urry, 1936; Evans and Goodman, 1941) has been the most common method for thorium analysis. Combined thorium and uranium data have been determined partly by direct radon ionization and thoron streaming (Keevil, 1938) and partly by indirect combination of radium determination and alpha counting (Keevil, 1944; Evans and Goodman, 1941). Recently, gamma-ray counting techniques have been utilized for the determination of uranium, thorium, and potassium (Evans and Raitt, 1935; Hurley, 1956; Adams et al., 1956; analytical part of this paper). Other workers have used combined gamma-beta counting (Wendt, 1955; Smith and

Flanagan, 1956). Coppens (1950) and others have used the autoradiographic technique to determine total alpha activity and also uranium and thorium contents of rocks.

The radiometric techniques, with the exception of some alpha counting, depend on calibration standards. These, according to Evans and Goodman (1941), were very poor prior to 1936; accordingly, analyses prior to this date are given less weight in this paper. Radioactive equilibrium must be assumed in order to calculate thorium and uranium contents from radiometric data.

Direct fluorimetric chemical analysis for uranium has recently been developed to a high degree of accuracy (Grimaldi, May and Fletcher, 1952; Adams and Maeck, 1954), and very recently chemical thorium analysis has been greatly improved (Grimaldi et al., 1954; and Grimaldi, unpublished).

PRESENTATION OF DATA

Data on uranium and thorium (when both are reported) and/or Th/U ratio shall be referred to in this paper simply as Th/U data (or values). Radium, uranium (alone) and/or radioactivity data will be referred to as "activity". These two types of data will be presented separately.

Th/U values are of direct importance to this study, therefore all such analyses of individual rocks which I

consider to be reasonably correct are included in table 1. A few older values which are not well validated, mainly those impugned by Evans and Goodman (1941), are excluded. Some of the values which have been presented, although reasonable, may not be correct. Wendt's (1955) combined beta-gamma counting method appears theoretically sound, but his data lack any checks by other methods or by other laboratories. Husmann's (1956) method is apparently similar to Wendt's, but it is not at all clear; his data also lack checks. Evans and Raitt's (1935) data, although accepted by Evans and Goodman (1941) and therefore purportedly interchecked, do not seem theoretically well based. Evans and Raitt used a combination of radium determination and total gamma count to calculate thorium indirectly. However, the gamma contribution from K^{40} was not considered; this should result in high thorium values. Their values are, in fact, among the highest in the table.

Summary data on Th/U in various geographical areas are presented in tables 2, 3, and 4. Estimates of world Th/U are given in table 5.

Activity is important in this study only in that it can help to isolate the levels of variability of uranium and thorium. This fact, together with the great bulk of the activity data, precludes its detailed presentation; therefore, only averages and summaries are presented in the

Table 1.- Th/U values for individual rocks. Arranged in order of increasing Th/U ratio.

LOCATION	ROCK	α /mg/hr	Thppm	Uppm	Th/U	REFERENCE
Wildschapbachthal	Granulite		2.7	7.9	0.34	Husmann, 1956
Massachusetts	Rockport granite	2.1	2.6	5.1	0.5	Evans and Goodman, 1941
"	Hbl'd granodiorite	0.34	0.45	0.82	0.55	"
Glen Cove, Colo.	Granite	2.8	4.3	6.5	0.7	"
Einetwald	Orthogneiss		3.7	3.9	0.94	Husmann, 1956
W. of Taschereau, Que.	Granitic type		0.9	0.58	1.5	Keevil, 1938
"	Same (replicate)		0.9	0.62	1.5	"
Blue Mt. Ont.	Syenodiorite		0.32	0.21	1.5	Keevil, 1944
Franklin, Maine	Granitic type		8.84	3.55	2.5	Keevil, 1938
"	"		8.84	3.72	2.4	Keevil, 1944
"	Granite	1.24	0.0	3.5	0.0	Evans and Goodman, 1941
"	Average		6.56	3.6	1.6	
Lake La Motte, Que.	Granite	0.64	2.04	1.25	1.6	Evans and Hatt, 1935
"	Granodiorite	0.81	2.5	1.6	1.6	Evans and Goodman, 1941
"	"		2.04	1.34	1.5	Keevil, 1944
"	Average		2.15	1.4	1.58	
Flohtgebirge	Granite		47.0	30.6	1.56	Wendt, 1955
Kerhino-en-Vannes	Gneiss		26.2	15.1	1.7	Coppens, 1950
Chelmsford, Mass.	Migmatite		7.3	3.4	1.7	Keevil, 1944
Zangershof	Orthogneiss		3.12	1.53	2.1	Husmann, 1956
Barryfield Quarry, Ont.	Granite				2.1	Keevil, 1944
Flohtgebirge	Granite		46.7	20.8	2.25	Wendt, 1955
Kingston, Ont.	Granitic type		3.1	1.4	2.3	Keevil, 1938
Massachusetts	Rockport xenolith	3.9	17.0	6.5	2.6	Evans and Goodman, 1941
Genanogue, Ont.	Granitic type		2.82	1.05	2.7	Keevil, 1938
"	Granite		2.71	1.10	2.5	Keevil, 1944
"	Average				2.6	

Table 1.- (Continued).

LOCATION	ROCK	α /mg/hr	Thppm	Uppm	Th/U	REFERENCE
E. of Tasohereau, Que.	Granitic types		4.4	1.59	2.8	Keevil, 1938
"	Granite		4.42	1.7	2.6	Keevil, 1944
"	Average				2.7	
Morbihan	Granulitic gneiss				2.75	Coppens, 1950
Quiberon	Granite				2.75	"
Bretagne	Bretagne granite				2.8	
Vassan Tp., Que.	Granodiorite		2.2	0.72	3.1	Keevil, 1944
Raymond, Calif.	Qtz monzonite	0.77	3.9	1.2	3.3	Finney and Evans, 1935
Quincy, Mass	Quincy granite		10.6	2.21	4.91	Keevil, 1938 (b)
"	"		12.4	2.78	4.5	"
"	"		11.7	3.55	3.32	"
"	"		10.2	3.67	2.78	"
"	"		9.0	3.46	2.6	"
"	"		7.0	2.7	2.59	"
"	"		7.8	3.1	2.52	"
"	"		5.9	2.94	2.0	"
"	"		18.7	5.1	3.7	Hee et al., 1954
"	"		10.4	2.1	4.9	"
"	"		10.6	3.05	3.7	Keevil, 1938
"	"		11.75	3.55	3.3	Keevil, 1944
"	"		9.3	3.01	3.1	"
"	"	1.7	8.1	2.7	3.0	Evans and Goodman, 1941
"	Average		10.3	3.14	3.42	
Schnecken felsen	Orthogneiss		11.0	3.3	3.4	Husmann, 1956
Cape Ann, Mass.	Granitic type		19.3	5.17	3.7	Keevil, 1938
"	Granite		17.1	5.05	3.4	Keevil, 1944
"	Average				3.55	
Ile-aux-Moines, (Morbihan)	Granulite				3.6	Coppens, 1950
Perris, Calif.	Lakeview tonalite		4.0	1.1	3.6	Keevil, 1944
Hampstead, N. B.	Granitic type		17.0	4.4	3.9	Keevil, 1938
"	Granite		17.0	4.63	3.7	Keevil, 1944

Table 1.- (Continued).

LOCATION	ROCK	α /mg/hr	Thppm	Uppm	Th/U	REFERENCE
Valverde, Calif.	Tonalite		5.0	1.3	3.8	Keevil, 1944
New Hampshire	Conway granite		67	12.7	5.27	Hurley, 1956
"	"		46.4	9.1	5.08	"
"	"		61.0	13.8	4.41	"
"	Average		32.0	10.3	3.2	
Copper Cliff, Ont.	Greighton granite	2.8	51.6	11.5	4.5	
Colorado	Mt. Manitou granite	3.2	17.0	3.5	4.8	Evans and Goodman, 1941
Massachusetts	Fitchburg granite	2.8	20.0	3.8	5.4	"
Fitchburg, Mass.	Granite migmatite		21	2.5	8.4	Keevil, 1944
"	Granite and peg.		19.0	3.1	6.1	Keevil, 1938
"	Average		10.1	2.5	4.2	
Winnipeg, Man.	Granite	0.36	2.7	0.34	6.2	Evans and Goodman, 1941
Red River, Man.	Granite		4	0.5	7.8	Keevil, 1944
Kaweah Quad, Calif.	Qtz monzonite	1.46	11	1.3	8.5	Finney and Evans, 1935
Rocky Hill Quad., Calif.	Granite	1.93	15	1.7	8.8	"
Kaweah Quad., Calif.	Granite	1.97	16	1.5	11	"
Milford, Conn.	NBS Milford granite		9.0	0.8	11.25	Hurley, 1956
Pikes Peak, Colo.	Red granite	5.77	48	4.1	12	Evans and Raitt, 1935 ^a
Churchill, Man.	Granite	1.00	8.7	0.62	14	"
Baileysbrom	Granulite		50	1.9	26	Husmann, 1956

¹ Keevil, 1938 (b) presents only Ra and Th data. Uranium and thorium/uranium ratios tabulated were calculated by my assuming radioactive equilibrium and using the relation $U = 2.84 \times 10^6$ where Ra is in $g \times 10^{-12}/g$.

² Evans and Raitt's data, although accepted by Evans and Goodman, may not be good since determinations are based on total gamma counting combined with uranium determination without considering the gamma contribution of K^{40} . This would result in high thorium figures and therefore in high ratios, which seems to be the case.

tables of activity. I have arranged the material to emphasize variation on several different levels or scales: table 2 presents data on variation of activity in individual plutons, tables 3 and 4 show distribution of activity among the members of several comagmatic series, and tables 5 and 6 present alpha activity of petrographic types in large geographic regions, culminating in estimates of world averages.

Discussion of distribution of Th/U values and activity in granitic rocks will follow the tables where they are available and will involve four levels of variation: 1) on hand specimen level (sampling homogeneity), 2) within pluton, 3) within comagmatic series, and 4) above the level of comagmatic series among main petrographic types and between major geographic regions. Correlation of activity and Th/U data with various geologic, chemical, and petrologic parameters will be considered last. Within each of these divisions activity data will be considered first, followed by consideration of Th/U data.

I Sampling Homogeneity

Sites of activity: Information on the location of radioactive elements in granitic rocks is fairly abundant, but the data are extremely variable and often contradictory.

This study will be concerned with the sites of activity in rock only in the effect that they might have on sampling homogeneity.

Total activity and Th/U are distributed differentially in the major rock-forming minerals, but estimates of the proportion of activity in these minerals vary widely (Larsen et al., 1956; Piggot, 1931; Hayase, 1955; Keevil, 1943; Coppens, 1950; Hee et al., 1954). Larsen et al. (1956) feel that most of the uranium in a rock is in the major rock-forming minerals.

Radioactivity is concentrated in the accessory minerals of granitic rocks. Larsen et al. (1956) estimate that 25 per cent of the uranium in a bulk rock is concentrated in the common accessories, such as zircon, sphene, and apatite. Coppens (1950) feels that 60-70 per cent of the alpha activity of granitic rocks is concentrated in accessories of all kinds. The Th/U ratios in accessory minerals are poorly known, but some recent excellent work has been done (Smith et al., 1957; Hurley and Faibairn, 1957). Common accessories are fairly evenly distributed and should not vitiate representative sampling. There have, however, been reports of very small, extremely active, sparsely and very irregularly distributed accessories such as monazite, xenotime, thorite, and uraninite (Deutsch and Picciotto, 1956). Larsen et al. (1956) feel that the

contribution due to such minerals is more than was formerly thought, but they do not attempt a quantitative estimate. Such sites of activity, if important, could easily complicate the sampling problem.

In 1950 Hurley found that the alpha activity of granites could be decreased by a factor of 5-10 by leaching in dilute acid. Subsequent studies have substantiated this discovery (Tauson, 1956). Brown et al. (1953) find that 40 per cent of the radioactive elements may be leached from a granite. Brown and Silver (1955) find that 15-38 per cent of the radioactive material in granitic rocks may be leached and find that 29 per cent of the uranium and 42 per cent of the thorium in the Essonville granite is leachable. Tilton et al. (1954) find 34 per cent of the uranium and 42 per cent of the thorium leachable in the same Essonville granite and conclude that the (rock) system was open with respect to thorium and its decay products. They feel that this leachable activity is in "chemically unstable and presumably interstitial phases of the granite." Some of the accessories, notably epidote, allanite, and apatite, may be significantly soluble in dilute acid (Neuerburg, 1956; Brown et al., 1953; Picciotto, 1950). Some investigators feel that thorium-rich allanite is a late hydrothermal mineral (Coulomb et al., 1956; Hutton, 1951).

Labile interstitial material or unstable hydrothermal minerals could have very irregular distributions in a rock body and could be concentrated in zones of shear and alteration (Coulomb et al., 1956); this would complicate any attempt at representative sampling.

Recent summaries of the location of uranium within granitic rocks (Neuerburg, 1956; and various papers by Larsen and Phair) tend to show that uranium is present in different sites of varying importance in different rocks. Thus, Larsen et al. (1956, p. 66) say: ".....not only the total uranium content but the amount of leachable uranium change in a roughly systematic way with rock composition and point to a predominantly petrogenetic control."

Empirical variation: The size of the sample which must be analyzed in order to represent accurately the radioactivity of a rock is a function of the distribution of the activity in that rock. However, from the foregoing discussion, it is obvious that the distribution is poorly known and apparently variable; therefore some empirical estimates must be considered. Billings and Keevil (1946, p. 180) conclude from their study of the White Mountain Magma Series, that "the specimens studied, which usually contained 1 to 10 cubic inches, must be smaller than the unit that would be necessary to obtain uniform values." Keevil et al.

(1943) find most determinations are moderately reproducible but that great variation is found in retests of individual samples. On the other hand, Evans and Goodman (1941, p. 463) find that "after a rock is ground, different 5 to 10 gram portions appear to contain equal radium contents within experimental uncertainty of measurement, which is estimated to be 8 per cent." The agreement on two samples of Quincy granite analysed by Hee et al. (1954) is notable when it is considered that they were done by the autoradiographic technique, a method which should be extremely sensitive to small scale radioactivity variations.

In all these cases, the amount of sample actually being tested is small; it is probable that a small increase in the size of the sample would greatly increase agreement between replicate analyses. Smith and Flanagan (1956, p. 316) find that "replicate uranium determinations of an analytical sample split from a previously obtained 40 lb sample of (Conway) granite gave results whose variance greatly exceeds (by a factor of about 10) the known variance of the analytical determination"; but they feel that "radioactivity determinations by beta counting should be relatively unaffected by sampling errors as the size of the sample for analysis is about 100 grams compared to a 1 gram sample for chemical analysis." This contention is borne out by their very homogeneous sampling (see table 7);

they find it feasible to detect small differences of uranium content in various facies of the rock with high statistical confidence.

There is very little data on variation of the Th/U ratio from sample to sample. A very rough idea of the variation in total activity may be obtained from the replicates in table 1. There is no compelling reason to suspect that the Th/U ratio should vary with total activity. In this regard it is notable that two samples of Fitchburg granite (Keevil, 1938, 1944) differ in thorium content by a factor of nearly two, yet their Th/U ratios differ by only 30 per cent.

Large differences in the activity of hand specimens may indicate a true sampling problem due to erratic sites of activity on a small scale (Smith and Flanagan, 1956); however, if the samples being compared are widely spaced, variation may also be due to areal variation within a given body. Such variation may be related to subtle changes in gross lithology. In some cases (e.g. Billings and Keevil, 1947) of claimed inhomogeneity of radioactivity, the area or volume over which homogeneity was sought is not specified; therefore the assignment of variation to a specific level is impossible. In most cases the lithology of samples was not closely examined.

II Distribution Within Plutons

Peripheral concentration of radioactivity within individual plutons has been observed by many authors (see table 7). Larsen et al. (1956) find approximately a sevenfold concentration of uranium in the border phase of the Boulder Creek batholith with respect to the interior. Davidson (1951) claims concentration of uranium in the outer portions of the alkali Nigerian granites. Slack and Whitham (1951) find a decrease in total gamma activity toward the center of both the Round Lake and the Elzevir batholiths in Canada. Radium is distinctly concentrated in the border phase of the Tanakami stock, Japan, according to Asayama (1954). Keevil et al. (1943) note a decrease in alpha activity toward the center of the Bourlemaque batholith. Ingham and Keevil (1951) in their study of the Elzevir, Cheddar, and Bourlemaque batholiths find that 6 out of 9 separate intrusions composing these bodies have peripheral concentration of alpha activity.

Vertical variation of radioactivity is apparently present within some granitic bodies. Soloviev (1936, p. 380) concludes from his examination of 52 samples from the Malka River laccolith that "the content of radium decreases systematically from the roof of the intrusive downward." Evans and Goodman (1941) have alpha counted two

Table 2.- Distribution of uranium and alpha-activity within individual plutons.

PLUTON	FACIES	NO	U ppm	α /mg/hr	RANGE	REFERENCE
Boulder Creek Batholith	Granodiorite				5.5-14.5	Larsen et al., 1956
"	Contact (basio) zone		9.3		3.4- 6.4	"
"	Intermediate zone		5.6		1.4- 3.4	"
"	Central (acidio) zone		2.4			
Teschereau, Ont.	Granodiorite					
"	Contact zone			0.31		Keevil, 1943
"	Central zone			1.01		
Bourlamaque Bath., Que.	Granodiorite (average)	150		0.37 ¹		Ingham, Keevil, 1951
"	Contact zone			0.7		"
"	Central zone			0.48		"
Cheddar Batholith	Granite (average)	175		1.85 ¹		Ingham, Keevil, 1951
"	Contact zone	28		3.01		"
"	Intermediate zone	93		1.97		"
"	Central zone	36		1.11		"
Elzevir Batholith	Diorite gabbro ²	12		0.29 ¹		Ingham, Keevil, 1951
"	Gray granite	86		0.65 ¹		"
"	Contact zone			1.72		"
"	Intermediate			0.72		"
"	Central zone			0.5		"
"	White granite	10		0.93 ¹		"
"	Contact zone			1.32		"
"	Intermediate zone			0.78		"
"	Qtz diorite	6		1.20		"
"	Contact zone			1.51		"
"	Intermediate zone			0.75		"
"	Syenite facies	17		1.3		"
"	Contact zone			1.86		"
"	Central zone			0.48		"
"	Red granite ²	1		1.56		"
"	Aplite granite ²	12		1.69		"

Table 2.- (Continued).

PLUTON	PACIES	NO. U ppm α /mg/hr	RANGE	REFERENCE
"Granite de Bretagne"	Average "granite" ²	215 4.9 ³	$\sigma = 0.19$	Coulomb et al., 1956
Malka River Laccolith	Granodiorite ⁴	5.37 ⁵		Soloviev, 1936
Mikumo Batholith	Normal granite ²	9 2.16 ⁵	1.2-3.5	Assayama, 1954
"	Biotite granite	2.44 ⁵	1.2-3.8	"
Tanakami Stook	Biotite granite	13 5.79 ⁵	3.3-9.2	Assayama, 1954
"	Contact type	7 6.5 ⁵	3.3-9.2	"
"	Central type	6 5.0 ⁵	4.2-5.3	"

¹ Weighted average.

² No significant peripheral concentration of uranium.

³ Value is arithmetic mean. Geometric mean is 3.4.

⁴ Peripheral concentration.

⁵ Calculated from radium, assuming radioactive equilibrium and using the relation $U = 2.84 \times 10^6 \text{ Ra}$.

samples of Quincy granite from the same locality. One sample (12A₁) is from the surface and emits 1.42 alphas/mg/hr, the other sample (12A₂) is from a depth of 600 feet and yields an average of 1.9 alphas/mg/hr. Hurley (1954) finds that the activity of the Quincy granite is higher near the surface (17.2 alphas/cm²-h at a depth of 3 feet) than at greater depths (8.8 alphas/cm²-h at depths of 12 and 200 feet). (It is also interesting to note that in the same study Hurley finds that in samples taken from great depth the amount of leachable uranium is relatively smaller compared with total uranium than at the surface.) All these data, with the exception of Soloviev's, are very inconclusive.

Peripheral concentration of uranium is lacking in many plutons. Asayama (1954) finds that the outer part of the Mikumo batholith, Japan, is not enriched in uranium. Ingham and Keevil (1951) find that 3 out of 9 intrusions in eastern Canada lack peripheral uranium concentration. From their study of 215 samples from 3 French granite massifs, Coulomb et al. (1956) conclude that there is no radial increase in tenor of uranium. Gottfried and Larsen (1954) find no uranium enrichment in the border phases of a Woodson Mountain granodiorite stock in the Southern California batholith. "There is no obvious relation between position within the main batholithic mass (of the

White Mountain magma series) and concentration of (Phair, 1956, p. 317).

Variation in activity has, in some studies, been qualitatively related to slight petrographic variations. Soloviev (1936, p. 391) finds, in addition to a variation in radium content, that "the content varies with the petrographic nature of the granodiorite". relatively greater number of high values are reported for muscovite granodiorite than in biotite granodiorite more in biotite granodiorite than in hornblende granodiorite. Keovil et al. (1943) link peripheral increase in activity with increase in amount of accessory minerals. Corbett et al. (1956) find that uranium is concentrated at the greatest mineralization.

The amount of variation within a pluton may be related to the overall composition of the body. Larsen et al. (1943, p. 349) find that "the largest variations are in the more active granitic rocks, smallest in less active basic rocks."

The average activity and degree of peripheral concentration may be related to the size of the pluton; however, observations are contradictory. Asayama (1943) finds marked peripheral concentration of radium in small Tanakami stock but none in the much larger Mt. Ingham batholith. Out of 9 plutons studied by Ingham and

(1951) the 3 smallest, and only these 3, lack concentration of alpha activity in their border zones; the largest plutons show the greatest contrast of activity between their centers and their borders. Gottfried and Larsen (1954) find no peripheral uranium concentration in the small Woodson Mountain granodiorite stock. Keevil et al. (1943) find that specimens from larger intrusions tend to be more variable. Phair (1956) reports that the average uranium content and the spread of uranium values in the Front Range, Pre-Cambrian, "Silver Plume" granite both increase with increase in the size of pluton; however, he notes that 3 ppm seems to be the lower limit in all bodies regardless of size. Phair (p. 317) also finds that, in the White Mountain magma series, "the biotite and amphibole granite of the main batholithic mass contain a larger proportion of uranium than corresponding granites of the outlying masses."

Variation of Th/U ratio within plutons is virtually unknown. Krylov (1956) has investigated the thorium and uranium content of two granite massifs. The thorium decreases peripherally in both; but the uranium increases peripherally in one, a stock, and is variable in the other, whose form is not stated. In both massifs the Th/U ratio apparently increases toward the borders. Studies of small granite stocks in Kirgizia, Russia, demonstrate " a slight

enrichment in the uranium content of the intrusion, but a concomitant impoverishment in thorium, along the contact zone" (Davidson, 1957, p. 8).

Summarizing, some plutons increase in activity toward their upper and outer parts; other plutons lack such systematic variation. The bulk of evidence seems to support higher total activity as well as greater peripheral concentration in larger and more acidic plutons. Th/U ratio may generally decrease peripherally, but documentation for this statement is very poor. Variations generally seem to be related to slight petrographic variation; however, it seems unlikely that the slight variations noted would be enough in themselves (i.e. barring the effect of position in the pluton) to account completely for the observed variation in activity.

III Distribution Within Comagmatic Series

The general increase of activity with decreasing age and increase in total alkali and silica within any given comagmatic series is a well known and well documented fact; therefore, it will not be developed here. Most of the authors cited in the last section have remarks and data on this subject. These are summarized in tables 2, 3, and 4.

Unfortunately, information on the distribution of

Th/U values within comagmatic series is practically lacking. Finney and Evans (1935) find that thorium increases relative to uranium between a quartz monzonite (Th/U 8.8) and a granite (Th/U 11) from Kaweah, Quadrangle, California (see table 1). Only two rocks are considered, and the difference is very small; therefore, the data are practically meaningless. Szadeczky-Kardoss (1953) presents ionic specific gravity as an explanation for what he seems to feel is a well-known fact: the enrichment of thorium over uranium in alkalic rocks. His data are not available.

IV Distribution in Rock Types Above the Level of Comagmatic Series

The purpose of this phase of the review is to attempt to ascertain the extent to which uranium and thorium are controlled by rock type. For example, the use of a term such as granite or quartz monzonite defines the amount of calcium or potassium in a rock within fairly definite limits. To what extent is this true of uranium and thorium and to what extent do they depend on the particular locality, the particular comagmatic series, or the particular conditions of genesis of the rock, independent of lithologic type? Some writers suggest that activity of petrographic types may be systematic within a given series but not

outside that series (Larsen et al., 1956; Larsen, 1954).

A rough idea of distribution of activity in petrographic types belonging to different comagmatic series may be obtained from figures 1, 2, and 3. Averages of the series may not be compared, since average compositions of different series are not the same. Petrographic types in the White Mountain magma series are obviously more radioactive, more uraniferous, and richer in thorium than corresponding petrographic types in other series; the White Mountain magma series, however, is extremely alkalic. Any given petrographic type in the other series is of roughly the same activity. For more detailed analysis the reader is referred to Billings and Keevil (1947), Larsen and Keevil (1942, 1947), and Larsen et al. (1956).

Keevil et al. (1943, p. 352) have made an extensive survey of the alpha activity of rocks from major geographic regions of North America. Their data are presented in table 5. They find that:

"Acidic intrusives in the Canadian Precambrian Shield show at least a fivefold variation, those near the western borders being more radioactive than those in Quebec and Ontario, which lie some distance within the Shield.

"Granites and granodiorites from southwestern Quebec have the lowest activity found for these types of rocks.

Table 3.- Distribution of uranium and alpha-activity in members of three magma series of the western U.S.

SERIES	MEMBER (type)	NO. SAMPLES	U ppm α /mg/hr	RANGE	REFERENCE
Idaho Batholith	Quartz diorite		1.8		Larsen et al., 1956
"	Granodiorite		2.3		"
"	Quartz monzonite		3.1		"
"	Muscovite granite		1.6		"
"	Fine granodiorite		1.0		"
"	Average		2.1		"
Sierra Nevada Batholith	Gabbro		0.6		Larsen et al., 1956
"	Quartz diorite		1.2		"
"	Granodiorite		2.4		"
"	Quartz monzonite		3.4		"
"	Muscovite granite		3.0		"
"	Average		2.3		"
Southern Calif. Batholith	San Marcos gabbro	11			Larsen, Keevil, 1947
"	Average gabbro		0.43		"
"	Average gabbro	10	0.3	0.2-2.3	Larsen et al., 1956
"	Bonsal tonalite		1.05*		Larsen, Keevil, 1942
"	Bonsal tonalite				Larsen, Keevil, 1947
"	Lakeview tonalite		1.39*		Larsen, Keevil, 1942
"	Green Valley tonalite	5	1.01		Larsen, Keevil, 1947
"	Average tonalite		0.8		"
"	Average tonalite	18	1.4	0.3-3.0	Larsen et al., 1956
"	Riverside granodiorite				Larsen, Keevil, 1942
"	Average granodiorite	13	0.65		Larsen, Keevil, 1947
"	Average granodiorite	37	1.8		"
"	Granite and Qtz-monzonite		2.0	0.7-4.8	Larsen et al., 1956
"		5	2.5		Larsen, Keevil, 1947
"		9	4.2	2.7-6.0	Larsen et al., 1956

* Calculated by me from radium figure of author, using relation $U = 2.84 \times 10^6 \text{ Ra}$ and assuming radioactive equilibrium.

Table 4.- Distribution of uranium and alpha activity in members of four New Hampshire
magma series.

SERIES	MEMBER (type)	NO. OF SAMPLES	U ppm	α /mg/hr	RANGE	REFERENCE
Highlandcroft Magma Series	Diorite and Quartz diorite	2	0.58	0.58- 0.59		Billings, Keevil, 1946
	Granodiorite	2	1.44	1.21- 1.68		"
Oliverian Magma Series	Quartz diorite	4	1.04	0.61- 1.60		Billings, Keevil, 1946
"	Granodiorite	6	1.35	0.59- 3.07		"
"	Quartz monzonite	6	2.46	1.29- 3.0		"
	Granite	11	2.67	1.29- 4.49		"
New Hampshire Magma Series	Quartz diorite	3	0.57	0.15- 0.85		Billings, Keevil, 1946
"	Quartz monzonite	16	1.84	0.94- 2.67		"
"	Lebanon granite	5	2.3	1.7 - 2.9		Evans, Goodman, 1941
	Average granite	18	2.55	1.29- 4.55		Billings, Keevil, 1946
White Mountain Magma Series	Gabbro and norite	4	1.10	0.56- 1.82		Billings, Keevil, 1946
"	Syenite	8		1.20-20		Larsen et al., 1956
"	Syenite	9	2.16	0.79- 5.03		Billings, Keevil, 1946
"	Quartz syenite	3		4.5 - 7.2		Larsen et al., 1956
"	Quartz syenite	9	3.09	1.6 - 5.12		Billings, Keevil, 1946
"	Quartz monzonite	2		1.8 - 7.6		Larsen et al., 1956
"	Granite porphyry	2		6.4 - 9.4		"
"	Amphibole granite	23		2.3 -23.0		"
"	Amphibole granite	6	3.26	2.12- 4.5		Billings, Keevil, 1946

Table 4.- (Continued).

SERIES	MEMBER (type)	NO. OF SAMPLES	U ppm	α /mg/hr	RANGE	REFERENCE
White Mountain Magma Series	Conway granite	16		4.56	1.59- 8.69	Hurley, 1956
"	"	1	9.1			"
"	"	1	10.3			"
"	"	1	12.7			"
"	"	1	13.8			"
"	"	17	11.7		4.3 -19.5	Larsen et al., 1956
	Conway granite					
	Red facies					
	weathered	12	55.5 ¹		-64	Smith, Flanagan, 1956
	fresh	12	57.2 ¹		-65	"
	Green facies					
	weathered	10	49.8 ¹		-52 ²	"
	fresh	10	42.1 ¹		-46	"

¹ Equivalent uranium.

² One anomalous figure of 82 ppm equivalent uranium is not considered. I have averaged it, however.

"Acidic rocks from California, British Columbia, Saskatchewan, Manitoba, Greenland, Newfoundland, Finland, and Tanganyika show intermediate values of activity, while Paleozoic igneous rocks along the eastern seaboard, intrusives in Colorado, some in Yellowstone Park and the Northwest Territories, show the highest content of radioactive elements, as determined by alpha-ray emission from the uranium, actino-uranium, and thorium series."

Contrasts were not so great in basic rocks as in the acidic rocks referred to here.

It is unfortunate that more petrographic data are not available on these rocks; classification such as granitic, intermediatic, and basic is very vague. If one were to compare the average potash-feldspar/plagioclase ratios of several of the "intermediate" groups one might find some very marked differences which might be related to differences in activity.

Activity of petrographic types from various parts of the world and average world figures are presented in table 6. Different units of measurement, slightly different classification terminology, and small numbers of samples make it very hard to obtain any meaningful idea of world variations or averages.

Information on distribution of Th/U in some rock bodies and rock types by geographic region is available. Some recent Russian work is shown in table 7.

Table 5.- Distribution of alpha-activity in North American rock types.

REFERENCE	LOCATION	ROCK TYPE	NO. SAMPLES	α /mg/hr	RANGE
Piggot, 1931	N.E. States	Granite	16	0.9*	
Keevil et al., 1943	Quebec, West	Granitic	7	0.42	0.08 -1.01
"	"	Intermediate	4	0.36	0.18 -0.4
"	"	Basio	6	0.43	0.15 -0.72
"	Manitoba	Granitic	11	1.04	0.18 -2.37
"	"	Intermediate	3	0.55	0.13 -0.91
"	"	Basio	5	0.46	0.00 -0.99
"	Saskatchewan	Granitic	3	1.13	0.88 -1.56
"	Southern and Central	Granitic	10	1.15	0.004-2.20
"	British Columbia	Intermediate	15	1.69	0.1 -3.4
"	"	Basio	8	0.37	0.21 -0.85
"	Newfoundland	Granitic	4	1.54	0.29 -2.31
"	"	Intermediate	4	1.24	0.65 -1.96
"	"	Basio	4	0.43	0.32 -0.52
"	N.E. of Yellowknife	Granitic	18	1.95	0.6 -3.77
"	N.W. Territory	Basio	10	0.27	0.7 -0.59
"	California and Nevada	Granitic	2	1.95	1.93 -1.97
"	"	Intermediate	9	0.84	0.00 -1.79
"	"	Basio	9	0.79	0.56 -1.46
"	Eastern States	Granitic	18	2.02	0.17 -7.82
"	"	Intermediate	1	1.19	
"	"	Basio	40	0.13	0.02 -2.25

Table 5.- (Continued)

REFERENCE	LOCATION	ROCK TYPE	NO. SAMPLES	α /mg/hr	RANGE
Keevil et al., 1943	Maritimes, N.B. and N.S.	Granitic Intermediate Basio	3 2 8	2.43 0.38 0.60	1.99 -3.30 0.34 -0.42 0.18 -0.89
"	Ontario, Sudbury Area	Granitic	2	2.72	2.5 -2.95
"	"	Intermediate	2	1.24	1.19 -1.29
"	"	Basio	10	0.82	0.31 -2.1
"	Yellowstone Park	Granitic	2	3.0	2.8 -3.2
"	"	Intermediate	3	1.48	1.15 -1.80
"	"	Basio	13	0.79	0.4 -1.6
Keevil, 1943	Bourlenuque, Quebec	Granodiorite	94	0.48	
Ingham, 1951	North America	Granodiorite	1000	1.4	

* This figure is not α /mg/hr but μ Ra/g $\times 10^{12}$, and it is high by a factor of 2, according to Evans and Goodman (1941).

Table 6.- Estimates of large areal activities, including world averages.

REFERENCE	LOCATION	ROCK TYPE	NO. OF SAMPLES	$\frac{g\text{ Ra}}{g \times 10^{12}}$	$\frac{U\text{ ppm}^1}{\alpha/\text{mg/hr}}$	RANGE
Asayama, 1936	Japan, Hiei Region	Granites	14	1.25	3.55	
Asayama, 1954	Japan	Granodiorite and Quartz diorite	12	0.72	2.04	
"	"	Quartz porphyry and Granodiorite	4	0.99	2.81	
"	"	porphyry	80	1.36	3.86	
"	"	Granite	12	1.85	5.25	
"	"	Aplite	108	1.33	3.78	
		All granitic rocks				
Keevil, 1943	Tanganyika	Granites	4		1.55	0.12-3.0
Keevil et al., 1943	South Africa	Granite	4		1.58	0.12-3.10
	"	Basic	11		0.96	0.27-1.64
Keevil, 1943	Finland	II. Post-Bothnian	2		1.63	1.13-2.2
Piggot, 1938 ^a	Finland	Granitic rocks				
"	"	I. Post-Swlonian	4	0.94	2.67	0.63-1.23
"	"	II. Post-Bothnian	8	2.52	5.20	0.91-5.51
"	"	IV. Rapakivi	9	1.53	3.13	0.92-2.74

Table 6.- (Continued)

REFERENCE	LOCATION	ROCK TYPE	NO. OF SAMPLES	g Ra/ g x 10 ¹²	U ppm ¹	α /mg/hr	RANGE
Jeffreys, 1936 ²	World average	"Granite"	122	2.38	4.91		
Asayama, 1954 ⁴	World average	"Granite"	212	2.02	4.17		
Evans and Goodman, 1941 ⁵		"Granite"	21	1.69	3.49		

¹ Uranium figures calculated from radium, assuming radioactive equilibrium and using the relation $U = 2.84 \times 10^6 \text{ Ra}$ (Ra in g Ra/g x 10²).

² Evans and Goodman (1941) indicate that Piggot's figures are high by a factor of 2.

³ Jeffreys' estimate is high.

⁴ Asayama uses Jeffreys' collection plus some new data.

⁵ Evans and Goodman present 21 superior analyses, almost all from North America.

Table 7. - Uranium and thorium in Russian granites
(From Davidson, 1957).

LOCALITY	NO. OF SAMPLES	FIELD RELATION- SHIPS	U ppm	Th ppm	Th/U	AUTHORITY
Ukraine	107	Basement batholith	5.7	33	5.8	Vol.* 7, 1956
Terskei Alatau Kirgizia	33	Batholith	3.7	9	2.4	"
Kirgizia	65	Four small stocks	4.0	14	3.5	"
Northern Karelia	11	Migmatite field	2.3	14	6.1	Vol.* 5, 1957

*Volume referred to by Davidson is the Russian publication
Trudy Radievegо Instituta im. V.G. Khlopina.

Data from Senftle and Keevil (1947) on the regional variation of alpha activity, thorium and uranium content, and Th/U ratios in intermediate and granitic rocks are summarized in table 8 and in figure 32. Note that both intermediate and granitic Th/U ratios, and to a lesser extent thorium contents, form linear trends when plotted against one another and against uranium. Central Canada is an exception to this; it seems to be depleted in thorium relative to uranium. Keevil et al. (1943) found this region anomalously low in activity.

Table 8.- Thorium and uranium contents and alpha-activity of rock types by geographic region (after Senftle and Keevil, 1947).

LOCATION	ROCK TYPE	NO. OF SAMPLES	α /mg/hr	Th ppm	U ppm	Th/U
Central Canada "	Granitic	126	2.68	11.2	4.68	2.4
	Intermediate	23	2.63	12.9	3.7	3.5
Ont. and Que. "	Granitic	396	1.72	8.89	2.90	3.06
	Intermediate	72	1.41	7.95	2.09	3.81
Southwestern states	Granitic	50	3.11	13.3	4.2	3.17
	Intermediate	10	1.87	11.4	2.52	4.53
Eastern states "	Granitic	143	2.46	11.0	3.4	3.23
	Intermediate	20	1.32	6.58	1.34	4.98
Northwest coast "	Granitic	92	2.4	12.2	3.52	3.47
	Intermediate	23	1.7	7.42	2.56	2.9
World, excluding North America	Granitic	30	3.03	15.2	4.35	3.50
Western states "	Granitic	52	2.01	11.1	3.1	3.59
	Intermediate	23	1.97	11.0	2.57	4.27
Central states "	Granitic	76	2.55	13.1	3.52	3.72
	Intermediate	17	1.89	10.6	2.2	4.82
Rocky Mountain states	Granitic	199	3.39	18.7	4.98	3.76
	Intermediate	52	2.35	13.0	2.72	4.80
Northeast coast "	Granitic	123	3.2	19.8	4.98	3.98
	Intermediate	31	2.25	8.86	3.88	2.33

Per cent standard deviations have been calculated for Senftle and Keevil's data and are presented in table 9.

Table 9. - Per cent standard deviation for Senftle and Keevil's regional variation data.

TYPE	α /mg./hr.	Th ppm	U ppm	Th/U
Granitic	19.9 %	26.2 %	19.5 %	13.1 %
Intermediate	22.3 %	18.3 %	29.8 %	23.2 %

Note that the variation of the Th/U ratios of granitic rocks is considerably less than the variation of total alpha activity, thorium, or uranium. This indicates that thorium and uranium must generally vary together; furthermore, the variation in Th/U ratio may be predominantly controlled by variation in thorium. In the intermediate rocks the reverse seems to be true; the variation of Th/U ratio seems to be controlled more by variation in uranium than by variation in thorium. There appears to be considerably less co-variation of thorium with uranium in the intermediate rocks than in the granitic rocks.

The figures have little absolute meaning because there is no assurance that, in comparing different regions of "granitic" rocks, Senftle and Keevil are actually comparing similar petrographic types; the relative values for thorium

and uranium, however, are significant because they are measured on the same rock types -- whatever they may be. Uranium is more variable than thorium in the intermediate rocks, but in the granitic rocks, thorium is more variable than uranium. An explanation for this fact is not readily apparent; some possibilities will be suggested in a later section.

Estimates of world Th/U values are presented in table 10. The classifications of rock types are very vague, and it is difficult to compare values presented by different authors. Those values published before 1936 are probably inaccurate.

CORRELATION

In this section, attempts at more specific, quantitative, and semiquantitative correlation between radioactivity or Th/U values and various other measurable properties of rocks will be summarized. The correlations fall almost entirely into two categories, chemical and mineralogical.

Potassium and radioactivity have commonly been found to co-vary in granitic rocks. As early as 1905, Strutt made the observation that radioactive elements were correlatable with content of potassium. Many other authors support this observation (Nier, 1938; Urry, 1936a; Larsen

Table 10.- Estimates of world Th/U ratio averages.

REFERENCE	YEAR	ROCK TYPE	NO. OF SAMPLES	Th ppm	U ppm	Th/U
Jeffreys	1936	Acid igneous	60	17.6		2.6
		Intermediate	4	18.9		3.6
		Basic igneous	77	7.8		2.7
Holmes	1937	Acidic igneous				1.7
Holmes	1938	Acidic igneous				2.5
Keevil	1938	Acid igneous				2.8
		Acid igneous				3.3
Nier	1938	Acidic igneous				3.3
Evans and Goodman	1941	Granite	9	10.3	3.8	3.7
		Acid igneous	26	13.0	3.0	5.0
		Intermediate	6	4.4	1.4	2.6
		Basic igneous	34	3.9	0.96	4.0
Evans, Goodman and Keevil	1942	Acid and inter- mediate igneous				4.0
Keevil	1944	Granitic rocks	19			3.35
Senftle and Keevil	1947	Granitic rocks	1287	13.35	3.96	3.39
		Intermediate	297	9.97	2.61	3.98

and Keevil, 1947). Hatuda (1936) finds that radium varies with potassium in Japanese granitic rocks. Asayama (1956) finds the same relation in samples from within the Hiei region, the Mikumo batholith, and the Tanakami stock. He also notes that the average radium content of the Mikumo batholith and of the Tanakami stock are roughly proportional to the average potassium content of each of the two bodies. Holmes (1930), using data from Poole and Joly (1924), finds that both radium and thorium increase with increasing potassium in three series of Finnish pre-Cambrian granitic rocks. Larsen and Keevil (1947), Larsen (1954), and Larsen et al. (1950), note the concomitant variation of uranium and potassium in the Southern California batholith; Larsen (1954) notes, in addition, the co-variation of equivalent uranium with potassium.

Some other writers report negative results in attempts to correlate radioactivity with potassium. Barth (1938), working with the same four Finnish pre-Cambrian series considered by Holmes (1936), finds that the radium content of samples from within two of the series lack any relation to their potassium contents. Billings and Keevil (1946) report that radioactivity does not vary with the potassium content of the White Mountain magma series, and that, in fact, some of the most potassium-

rich members of the series (syenites, etc.) are of abnormally low activity. Coulomb et al. (1957) find that uranium and potassium are not statistically correlatable in the 3 French granitic massifs which they examined.

Radioactivity generally increases in the later stages of differentiation; therefore, a correlation with silica might be expected. Asayama (1954) finds just such a correlation in the Hiei granite and in the Mikumo batholith. The Tanakami stock (Asayama, 1936), however, lacks such a correlation. Larsen and Keevil (1947) find approximate correlation of alpha activity and silica content in the Southern California batholith. Larsen and Keevil (1947), Larsen (1954), and Larsen et al. (1955) find correlation of uranium with Larsen's differentiation index ($1/3 \text{ SiO}_2 + \text{K}_2\text{O} - \text{FeO} - \text{MgO} - \text{CaO}$).

Some investigators have correlated radioactivity with petrography or mineralogy (Barth, 1938; Evans and Goodman, 1941). Barth (1938) finds very good correlation of radium and biotite content in two series of pre-Cambrian Finnish granitic rocks (the Svecofennian zone granites and the Rapakivi granites). Keevil (1943) finds a somewhat similar correlation in a heterogeneous group of rocks; as total alpha activity increases the ratio of activity

of the mafics divided by the activity of the felsic minerals also increases.

Increasing radioactivity has commonly been ascribed to increasing quantities of various accessory minerals. Keevil et al. (1943) prepared a graph of alpha activity versus amount of accessories (chiefly zircon and apatite). They find a strong positive correlation, but the estimate of the amount of accessories is only semiquantitative; therefore, such a correlation is of doubtful significance. In the Malka River laccolith, Soloviev (1936) finds that increasing amounts of radium are accompanied by increasing amounts of zircon. There is a concomitant increase in zircon and particularly in allanite with increasing radioactivity in the White Mountain magma series (Billings and Keevil, 1946). Asayama (1954) correlates radium with the number of pleochroic haloes in nine granites from the Tanakami-Mikumo area.

Ingham and Keevil (1951), in reference to the Cheddar batholith state that "no general relation was noted between color, composition, or texture of the pink granite (of the Cheddar batholith) and its radioactivity, although gneissic granites showed a greater proportion of high values."

Coulomb et al. (1956) find roughly log-normal distribution of uranium in six regional subdivisions of

215 samples from three French granite massifs.

In his preliminary study of the Silver Plume granites of the Front Range, Phair (1956) presents a semiquantitative graph showing increase of spread of uranium values with increase in the size of pluton.

Keevil (1938) finds that six Paleozoic and six pre-Cambrian granitic rocks contain respectively 3.3 and 2.3 ppm uranium.

ANALYTICAL METHOD: GAMMA-RAY SCINTILLATION

GENERAL THEORY

I Nature and Origin of Gamma-Rays

Gamma-rays are high frequency electromagnetic radiation originating in the atomic nucleus. They differ from X-rays only in origin; X-rays emanate from the electron shells of an atom. Radioactivity processes are known to produce gamma-rays ranging in energy from 10 kev to 7 Mev.

The energy and origin of gamma-rays may be related to the shell structure model of the nucleus. According to this model the nucleons composing a nucleus have energetically favored regions of existence analogous to the shell structure of atomic electrons. When this structure is disturbed for any reason the nucleons tend to revert to their original ground state (or energetically favored state) with emission of gamma-rays. The excited states of the nucleus are called isomeric states and the changes from one state to another, isomeric transitions. The shell structure model permits the prediction of possible isomeric states.

When an unstable heavy nucleus decays by alpha or beta emission the resulting nucleus may be left in any one of several possible isomeric states or in its ground state

(see figure 1). The frequency of decays to the various possible isomers is controlled by the stability of these isomers. Therefore, since radioactive decay is random, the relative number of decays to each isomer is, on the average, constant. The transition from any isomeric state to ground state follows almost immediately after the formation of the isomer and may occur directly or may occur indirectly through several other intermediate isomers. The frequency of various isomeric transitions is also controlled by stability relations of the isomers; therefore, the probability of a given transition is fixed, and its relative frequency is, on the average, constant. Isomeric transitions are accompanied by emission of gamma-rays whose energies depend on the magnitudes of the particular transitions which produce them. Therefore the relative frequencies of gamma-rays of different discrete energies are, on the average, constant.

If an atomic species decays by alpha emission to two or more isomers of the same new nuclide, the energies of the various alpha particles differ by approximately the energy difference between the isomeric states, and, therefore, by approximately the energies of the gamma-rays which are emitted in the transition from the higher to the lower energy isomers.

Figure 1.- Energy level diagram for Ra^{224} as obtained from the observed alpha and gamma radiations of Th^{228} . All energies are in Mev. The alpha particle energies are the kinetic energies, not total disintegration energies.

Beta decay is slightly more complicated and not well understood. The energies of beta particles for a given decay are not discrete but have a range of values. Nevertheless, the energy of a gamma-ray associated with a specific beta decay is discrete. Beta emission of the normal type may be considered as the transformation of a nucleon from the neutron state to the proton state,

freeing an electron and a neutrino. The new nuclide formed may be in an excited state, in which case it reverts to ground state by the emission of a characteristic gamma-ray.

Electron capture is a special type of beta decay in which an orbital electron, generally from the K-shell, is captured by the nucleus forming a proton from a neutron with corresponding emission of a positive beta particle (positron) and a neutrino. The nucleus formed may be in an excited state, in which case it emits characteristic gamma-rays during isomeric transition. A host of X-rays are formed by rearrangements in the electron shell consequent to the loss of a K-electron. Electron capture occurs in the decay of K^{40} to A^{40} and is of great geological importance.

II Interaction of Gamma-Rays with Matter

Gamma-rays interact with, and are absorbed by, matter in three principal ways: photoelectric absorption, Compton effect, and pair formation (Fano, 1953; Faul, 1954; Friedlander and Kennedy, 1956).

Photoelectric absorption: Photoelectric absorption may be thought of as the interaction of an electromagnetic wave with a single atom. The entire energy of the gamma-ray is transferred to an atomic electron which

is expelled from the atom with the energy of the gamma-ray minus the binding energy of the electron. Thus a discontinuous energy spectrum of photo-electrons results from a continuous energy spectrum of gamma-rays and is controlled by the levels of the binding energies of the K, L, etc., electrons. The atom which absorbed the gamma-ray is in an excited state, having a vacant electron shell, and a series of lower energy electromagnetic radiations are emitted in the region of X-ray and visible light as the electrons fall into lower shells. The electron which was expelled at high energy also causes excitation and produces lower frequency radiation in other atoms.

The rate of photo-electric absorption is a function of gamma energy and of the atomic number of the absorber. For high energy gamma-rays the rate of absorption falls off roughly as $E_{\text{gamma}}^{-7/2}$; for low energies the rate falls off more slowly and approaches E_{gamma}^{-1} . The absorption is roughly a function of Z^5 . The photo-electric effect is unimportant above 1 Mev except in the heaviest elements.

The Compton Effect: The Compton effect, or Compton scattering, may be thought of as a series of elastic collisions of a photon of gamma energy with a number of atoms; with each collision the photon is deflected from its original path and degraded in energy. This energy is

taken up in an atomic electron and results in emission of low-energy secondary electromagnetic vibration similar to the photo-electric effect. The total amount of low-energy radiation is equivalent to the amount of energy lost by the gamma photon in the absorber.

Absorption due to the Compton effect is a function of the energy of the gamma photon and, to a lesser extent, of the atomic number of the absorber. For gamma energy above 0.5 Mev the absorption falls off about as E_{gamma}^{-1} ; thus the rate of decrease of absorption with increase in energy of gamma-rays is much lower than in the case of photo-electric absorption. The Compton scattering per electron is independent of atomic number; therefore, the scattering coefficient per atom is proportional to atomic number only because of increasing number of electrons.

Pair Formation: If the energy of a gamma-ray is more than 1.02 Mev, the energy equivalent of the rest mass of two electrons, it may be absorbed by pair formation, the creation of a positron and an electron in the vicinity of the atomic nucleus. The positron so formed will generally combine immediately with an atomic electron, forming two quanta of annihilation radiation, and disappear. The electron which is formed has half the energy of the incident gamma minus the rest-mass

energy of the electron and is absorbed with the emission of secondary low-energy electromagnetic vibration. This mechanism of absorption is important only at high energy levels.

The Compton effect should account for the great bulk of the absorption of moderately energetic gamma-rays in the range 0.5 to several Mev, despite the fact that absorption is exceedingly complicated in minute detail due to the combination of the three effects. This implies that total absorption in this energy range is approximately proportional to atomic number (very roughly proportional to density) and inversely proportional to gamma-ray energy. Empirical absorption curves (Motteff, 1955) generally support this approximation.

The total secondary radiation emitted in these processes, with the possible exception of some pair formation, is equal to the total energy of the incident gamma-rays but at lower frequency.

The absorption, excitation, and secondary emission, even in Compton scattering, occur in an extremely short period of time, depending on the nature of the absorber but generally less than a microsecond (Kallman et al., 1952).

Some transparent crystals are composed of atoms which have electron shells with energy configuration

such that they emit secondary radiation partly or entirely within the visible range of light when irradiated with gamma-rays. Such secondary emission is called scintillation. Each incident gamma-ray results almost instantaneously in a pulse of light whose intensity is proportional to the energy lost by the gamma-ray in the crystal. Such pulses of light may be transformed by a photomultiplier tube into electrical impulses which are proportional to the intensity of the scintillations.

The spectrum of intensities of scintillations which is obtained from a mono-energetic source of gamma-rays is controlled by the way in which the gamma-rays are absorbed. If the photo-electric effect is dominant then the spectrum will be a unique one; that is, all the scintillations will be of the same intensity because all the gamma-rays either lose all of their energy in the crystal or lose none of it; there is no partial loss of energy with photo-electric absorption.

If absorption is by Compton scattering, however, a portion of the gamma-rays will give up only part of their energy in the crystal so that the scintillations they cause will be of lower intensity than the maximum, true estimate of gamma-ray energy. The spectrum will have a maximum at true gamma-ray energy, the photopeak, and a spread of values of lower intensity, the so-called

Compton spread or Compton scatter. The relative number of scintillations of maximum intensity and those of lower intensity will be a function of the density of the crystal, the size of the crystal, and the energy of gamma-rays, since these are the factors which control the amount of absorption.

If absorption is primarily by pair formation then the positron which is so formed immediately combines with an atomic electron to form two gamma-rays of 0.51 Mev each, annihilation radiation. If both these gamma-rays are captured in the crystal, then the pulse will be equivalent to the true value of gamma energy; if one or both of these gamma-rays escape, the scintillation formed will be accordingly less intense.

In the range of energy with which this paper is concerned, Compton scattering is by far the most important type of absorption; therefore a spectrum of a mono-energetic source will show strong Compton spread. It is obvious that, other factors being constant, the larger the crystal the smaller will be the Compton scatter component. Figure 2 is the spectrum of the mono-energetic K^{40} gamma-ray (1.47 Mev) which Hurley (1950) obtained with a thallium-activated sodium iodide crystal 1 3/4 by 2 inches. Figure 8 is the same spectrum obtained with the present crystal, which is 3 by 3 inches.

The amount of Compton scatter is important because, in measuring natural substances which are complex multi-energetic gamma emitters, the lower energy gamma peaks tend to be obscured by Compton scatter contribution from the higher energy emitters.

III Natural Gamma-Ray Emitters

Natural alpha-, beta-, and gamma-ray emitters are concentrated in the large unstable nuclides; particularly in three major decay series, Th²³², U²³⁵, and U²³⁸. Other natural emitters are summarized in the following table (table 11).

Iso- tope	type of disinte- gration	Half life in yrs.	Elements g/ton in ign. rx.	relative isotopic abundance	isotope g/ton in ign. rx.
K ⁴⁰	E.C., $\gamma_{1.47}, \beta$	1.2×10^9	25,900.	0.012	279.0
Rb ⁸⁷	beta	6.2×10^{10}	310.	27.8	8500.
In ¹¹⁵	$\beta, \gamma_{0.335}$	6×10^{14}	0.1	95.8	9.5
La ¹³⁸	E.C., $\beta, \gamma_{0.9}$	2×10^{11}	18.3	0.089	1.63
Nd ¹⁴⁴	alpha	5×10^{15}	23.9	23.9 (22.5)	570.
Sm ¹⁴⁷	alpha	1.3×10^{11}	6.47	15.1	97.9
Lu ¹⁷⁶	$\beta, \gamma_{0.31}, \text{others}$	4.6×10^{10}	0.75	2.5	1.95
Re ¹⁸⁷	beta	5×10^{10}	0.001	62.9	0.063
Pt ¹⁹⁰	alpha	10^{12}	0.005	0.0.2	0.0006

Table 11. - Natural radioactive isotopes. Largely from Friedlander and Kennedy (1956, p. 15 and appendix). Abundance data from Rankama and Sahama (1950). Isotopes in g/ton of igneous rock is my calculation. E.C. and γ_e stands for electron capture. Parentheses include other estimates.

Figure 2.- Gamma-scintillation spectrum for potassium.
(From Hurley, 1956).

The production of gamma-rays is inversely proportional to the half life and directly proportional to the abundance of the parent isotope. It is obvious that K^{40} is the most important contributor in most rocks. The long half life of indium effectually eliminates it from consideration. The gamma-ray production from lutecium must be very small, but in any case the energy of its gamma-ray is too low to interfere with any measurements involved in this investigation. The lanthanum gamma-ray is of the right order of energy but its half life is large and its abundance small compared to K^{40} , so it may be ignored.

The isotopic abundance of U^{235} is only 0.71 per cent of elementary uranium abundance; therefore it will not be discussed with the main thorium and uranium series. Its small contribution will be taken into consideration automatically in calibration methods to be described later.

Th^{232} decays to Pb^{208} through a series of 11 alpha or beta emissions, and U^{238} decays to Pb^{206} through a series of 14 alpha or beta emissions. Many of these decays are to one or more possible isomeric states whose transitions to isomeric states of lower energy produce characteristic gamma-rays. The gamma-rays associated with these series are summarized in table 1 of Hurley (1956).

If, for each decay of a parent nuclide, there is, on the average, a corresponding decay of each of its daughter products, then the series is said to be in secular radioactive equilibrium. Starting with pure uranium or thorium the decay will progress in such a way that equilibrium is approached at an ever decreasing rate. According to Hurley (1950, p. 396), "in the uranium series, 95 per cent of equilibrium is established in 3.5×10^5 years; in the thorium series only 30 years is required." Under equilibrium conditions all the members of the series are present in proportion to their half lives. The relative proportions of the daughter products are thus constant, and the ratio of one to another of the members of the series, or of any one to the parent uranium or thorium of the series, is fixed. It follows then that a measure of the amount of any daughter product will be an indirect measure of the amount of parent uranium or thorium.

Gamma-rays of various discrete, characteristic energies associated with a given decay are formed by isomeric states of fixed probability of formation; therefore, the ratio of the energies of these gamma-rays, is fixed and their number is proportional to the number of atoms of daughter product which produced them. Thus, measurement of the number of gamma-rays of any of the

characteristic energies is an indirect measure of the number of atoms of daughter product present. Furthermore, since the amount of any daughter product in an equilibrium series is controlled by the amount of parent uranium or thorium, the amount of gamma radiation of any discrete energy is likewise proportional to, and therefore a measure of, the amount of parent uranium or thorium. In practice, as will be seen, it is not feasible to measure the number of gamma-rays of discrete energy even for pure uranium or thorium because of the Compton spread from higher energy members of the series. Furthermore, in natural sources, uranium, thorium, and potassium are generally all present, further complicating the spectrum. Spectra of uranium, thorium, and K^{40} are shown in figures 5, 7, and 8.

K^{40} is an unstable, naturally occurring isotope which decays to Ca^{40} by beta emission and to A^{40} by electron capture. The decay is pictured in figure 3. About 12 per cent (Ahrens, 1954; Wasserburg, 1954) of the K^{40} decay is by electron capture to an isomer of A^{40} which emits a 1.47 Mev gamma-ray upon transition to its ground state. The isotopic abundance of K^{40} is constant in naturally occurring potassium (Nier, 1950; Rankama, 1954; Ahrens, 1954) and is approximately 0.0119. A measure of the amount of 1.47 Mev gamma emission is a measure of the

total amount of potassium present.

Figure 3. Decay scheme of ^{40}K to ^{40}A and ^{40}Ca . Subscript m indicates isomeric state, E.C. indicates electron capture.

IV Statement of Principle

Naturally occurring rocks contain uranium, thorium, and potassium in varying amounts; the measured spectrum from such a source will be a complex composite of all the gamma-ray energies in these series and their lower energy Compton scatter. The number of gamma emissions per unit time (the counting rate) at any given energy below 1.47 Mev has a uranium, thorium, and a potassium component. If the counting rate at each of three energy levels be measured for a pure uranium, a pure thorium, and a pure potassium standard to determine counting ratios,

and if the same three energy levels be measured for a compound source, then three equations may be formulated to solve for the three unknown counting rate components. These counting rate components may then be calibrated by comparison with counting rates of the pure standards if the amounts of thorium, uranium, and potassium in them are known. This problem is dealt with by Hurley (1956). If two of the energy levels considered are above 1.47 Mev then the equations may be considerably simplified since they will have no potassium component. The two high energies may be solved by a simple quadratic equation, and by utilising counting rate ratios these components may be stripped out of the lower counting level to give the third value (potassium). Furthermore, if the energy levels counted are chosen so that they are at the energy of a specific abundant gamma-ray in one series and at low counting rates of the other two, then the accuracy and precision of the method may be greatly increased.

INSTRUMENTATION

The gamma-ray spectrometer consists of three main components: the detector; the main electronic spectrometer, composed of several units; and the scaler.

I Detector

The specially constructed detector (see figure 4) consists of a 3 by 3 inch, cylindrical, thallium-activated sodium iodide crystal encased in aluminum. One base consists of a fused silica "window" which is in silica-gel optical contact with the three-inch-diameter photosensitive face of a DuMont model 6363 multiplier photo-tube. This photo-tube has a ten stage multiplier with optimum operation between 1000 and 1400 volts. It has been operated with a 1270 volt potential, giving a total amplification of approximately one million. The tube has maximum response to scintillations of $4400 \pm 500 \text{ \AA}$. The photomultiplier tube is shielded from magnetic forces by a mu-metal shield supplied by the DuMont company. A bleeder assembly at the base of the photo-tube distributes the voltage to the dynodes. No pre-amplifier is needed to send the signal through approximately 8 feet of coaxial cable to the spectrometer.

The crystal is designed to be used with an annular cannister and is therefore exposed on all sides not in contact with the photomultiplier. Sufficient room is present, however, at the top of the crystal chamber for small cylindrical source cans; such a can has been drawn on the diagram. The entire detector assembly may be removed from the main part of the shield.

Figure 4.- Gamma-ray detector and shield. Steel plates are one inch thick.

The detector shield is constructed of stacked, one-inch, steel plate with holes suitably cut to house the cylindrical liner, the detector probe, and the six-inch solid plug which covers it. A minimum of six inches of steel shields the crystal; this eliminates almost all external radiation since the tenth-value thickness of steel is less than four inches. Scant activity which may enter along the contacts of the steel plates is largely eliminated by a cylindrical liner and the outer case of the detector assembly, which are approximately one inch in combined thickness. Furthermore, the detector is situated in a four-foot deep concrete pit with several feet of concrete on all sides. Steel was used rather than lead because it has lower inherent radioactivity.

II Spectrometer

The main spectrometer unit, the commercially available Tracerlab Advanced Spectrometer Model RLP-3, with some minor deletions of unnecessary components, is composed of three main units: Precision High Voltage Supply (RLI-7), Pulse Height Analyzer Amplifier (RLX-12), and a Pulse Height Analyzer (special). The high voltage supply and the amplifier are connected to line current through a Sola Constant Voltage Transformer to stabilize the amplification. This is particularly necessary for the high voltage,

because the amplification in the multiplier photo-tube varies roughly as the seventh power of the voltage potential. The high voltage may be adjusted in steps of ten volts up to 1500 volts. Once changed, the high voltage is very difficult to reset precisely.

The Pulse Height Analyzer Amplifier is composed of two units, the Linear Amplifier Subchassis (RLA-1) and the Widener and Gate Generator (RLA-3). The linear amplifier increases the pulse which comes to it directly from the detector by a factor variable up to 8100. This so-called "gain" is controlled by two rotary switches, the fine gain and the coarse gain, which multiplicatively combine amplification factors of 90. The fine gain control attenuates the input signal in 0.1 steps from 2:2 to 1.2:2. The coarse gain permits attenuation of the signal by steps of 2:1 from 1:1 to 32:1. At a setting of 16 on the coarse gain and 1.6 on the fine gain the total gain is 1500.

The Widener and Gate Generator Subchassis shapes the pulses so that they will be suitable for pulse height analysis in the next step.

The Pulse Height Analyzer is a discriminator unit; it is used to determine the number of shaped pulses of various amplitudes (voltages) coming from the Widener and Gate Generator Subchassis. The amplitudes of the pulses

which can be discriminated by the unit vary from 3 to 105 volts. Elimination of pulses below any voltage in this range is accomplished by the use of an externally controlled, ten-turn potentiometer called the threshold. The threshold dial is calibrated from 0 to 1000 corresponding to from 3 to 105 volts; each division is equivalent to 0.1 volt. Another discriminator, also a ten-turn potentiometer, the "window", eliminates pulses above a certain voltage. This control is variable from 0 to 100 volts and has a dial reading from 0 to 1000, as on the threshold. This upper discriminator is arranged so that it "rides" on the threshold control giving in effect an adjustable "window" of voltages through which pulses of the proper voltage may pass, but above and below which all pulses are eliminated.

The pulses which pass through the window are recorded on a Baird Atomic Model 131 Cold Cathode Glow Tube Scaler. This is a decade scaler in which the units digit counts on a five microsecond decade strip; the next two digits, tens and hundreds, count on deca-tron glow tubes; and the next figures register on an electro-mechanical counter.

The entire spectrometer and detector are situated in a temperature- and humidity-controlled room; ambient temperature varies from 72 to 76 degrees and relative

humidity from 45 to 50 per cent.

The components of the spectrometer operate together in the following way. Gamma-quanta impinge on the sodium iodide crystal resulting in scintillations of different intensities corresponding to the energy of the gamma-rays. These scintillations activate the photo-sensitive surface of the photomultiplier tube causing emission of a number of photo-electrons proportional to the intensity of the scintillation. These photo-electrons are linearly amplified about one million times across ten dynodes of the multiplier photo-tube resulting in a low-voltage signal pulse. This signal pulse is further amplified approximately 1500 times in the linear amplifier. Variation in the total amplification of the signal pulse, due either to the high voltage or to the linear amplifier, varies the pulse height (voltage) corresponding to a gamma-ray of given energy; variation of amplification will expand or contract the entire spectrum. The pulse height analyzer can measure pulses from 0 to 100 volts; therefore, pulses which are amplified too much will be above the range of the analyzer. By lowering the amplification high energy gamma pulses may be brought within the range of the analyzer.

The threshold adjustments control the position of the "window" within the measurable spectrum. The width

of the window may be varied from 0 to 100 volts and placed anywhere from 3 to 105 volts. The specific relation between the pulses and the intensity of gamma-scintillation is controlled by the amplification.

If the total amplification is made constant, the window may be set at some arbitrary width, generally several volts, and may be moved in discrete jumps, by threshold adjustment, across the entire 102 volt range, and the number of signal pulses per unit time at each position may be counted. In this way a spectrum of number of pulses per unit time of different amplitudes (voltages) may be made. The amplitudes bear a linear relationship to the intensity of scintillations in the detector crystal and also, therefore, to the energy of the gamma-rays which caused those scintillations. The specific value of that linear relationship depends on the total amplification and may be simply calibrated by using gamma-sources of known energy.

OPERATION

I Calibration

In order to put the spectrometer into operation it is necessary to calibrate the threshold units in terms of gamma-ray energy for a given amplification. Sources of known gamma-ray energy are used to set up a calibration

curve between gamma-ray energy and threshold value. The position of the photopeak of a standard of known energy is located by moving the "window" in small increments across the peak. Numerous natural and artificial standards were counted and the positions of their peaks plotted to give the calibration curves in figure 5. Two curves are shown in this figure to indicate the effect of change in amplification on calibration. The position of the peaks in the spectrum may be changed by varying the amplification. In curve 2 of figure 5 the 0.661 Mev Cs^{137} peak is set at 210 threshold units; the highest energy gamma-ray from the thorium, uranium, or potassium series (the Th^{208} 2.62 Mev peak) then falls in the upper part of the spectrum at 840 threshold units. The spectrum is spread as much as possible while keeping the highest energy peaks in the measurable range. Curve 2 is the final calibration curve, made with operational amplification; one threshold division is equivalent to 2.9 kev of gamma-ray energy.

The resolution of the instrument is defined as the half-peak width divided by the distance of the peak from the origin (Friedlander & Kennedy, 1956). The resolution has been calculated for different peaks and amplifications giving varying results; generally better resolution (lower figure) is found at higher energy. At operational

amplification the resolution of the Cs^{137} standard is 9 per cent.

II Spectra of the natural series

Spectra of the uranium and thorium series and of potassium are presented in figures 6, 7, and 8. The uranium and thorium standards used to obtain these spectra were prepared by the New Brunswick Laboratory of the U.S. Atomic Energy Commission. The uranium standard is uraninite diluted with dunite and is free of thorium; the thorium standard is monazite diluted with dunite and carries a small amount of uranium. The potassium standard is a large annular cannister filled with reagent grade K_2CO_3 . The spectra were obtained using a 20 unit window (2 volts or 53 Mev) moved in increments of 20 threshold units (2 volts or 53 Mev).

The relative positions of the three spectra are shown superimposed on each other in figure 9. The relative form of any one spectrum, peak to valley and peak to peak, is a function of the size of the crystal (compare spectra to Hurley's spectrum in figure 10) and is therefore constant. The absolute height of the peaks and valleys is a function of the activity of the source; and, therefore, the relative heights of peaks of uranium to thorium to potassium are not significant

Figure 5.- Calibration curves for the gamma-ray spectrometer.

in this figure. The position and relative isolation of the peaks is of importance. Change of source geometry, barring a thick source with high self-absorption, will not effect the relative position or discreteness of the peaks.

III Choice of Counting Channels

Counts must be made with the "window" set at three gamma-energy levels, here called channels, in order to solve for the three unknown counting rates (uranium, thorium, and potassium). A fourth channel is counted to act as an independent check on the uranium and thorium. Potassium does not contribute counts to channels above 1.47 Mev, and uranium contributes only a relatively small number of counts above 1.75 Mev. From observation of figure 9, it is apparent that the Tl^{208} , 2.52 Mev peak in the thorium series; and the Bi^{214} , 1.76 Mev peak in the uranium series are relatively discrete and free from potassium contribution. The higher energy peak is also almost free from uranium contribution so that it is a very efficient thorium channel. The uranium 1.75 Mev channel, although not so efficient as the thorium channel, is free from potassium and has a relatively small contribution from thorium compared to lower energy uranium peaks; therefore it is chosen as the uranium channel. The choice of the potassium channel is obviously the

Figure 6.-- Gamma-ray scintillation spectrum for pure uranium series in secular radioactive equilibrium.

Figure 7.- Gamma-ray scintillation spectrum for thorium series in secular radioactive equilibrium.

Figure 8.- Gamma-ray scintillation spectrum for K_2CO_3 .

Figure 9.- Compound spectrum. Uranium, thorium, and potassium spectra superimposed.

Figure 10.- Gamma-scintillation spectrum for thorium series in equilibrium. (From Hurley, 1956).

1.47 Mev peak, since K^{40} is mono-energetic.

A fourth channel is counted as an independent check on the high-energy, 1.76 Mev, uranium channel. The uranium 1.12 and 0.61 Mev peaks are prominent possibilities. The 1.12 Mev peak is more efficient than the lower energy peak, but the counting rate at the lower energy is so much faster that greater precision may be attained at this energy in a shorter period of time. The contribution due to thorium is relatively higher at the low level making this channel a good independent check on the high energy thorium peak as well. The variation of counting rate with normal expected amplification drift was tested, as explained in the paragraph on choice of width of operational window; it was found that the low energy channel was more stable than the 1.12 Mev channel. For these various reasons, the 0.61 Mev channel was chosen as the fourth channel.

IV Width of Operational Window

Three criteria were utilized in the choice of window width: first, the count to background ratio must be as high as possible, where background is considered to be true background plus counts due to the components not being measured; second, the total counting rate must be as high as possible; and third, the variation in counting rate caused by normal expected drift in

amplification must be minimized.

By observing the 1.47 Mev peak in figure 9 it may be seen that with increasing window width, past a certain point, the number of counts due to potassium increases at a much lower rate than does the number of counts due to both uranium and thorium (background in this case); thus the count to background ratio decreases as window width increases. It might appear that the narrower the window the better; however, time considerations demand that the counting rate be as high as possible; and, therefore, an intermediate width must be used. By visual observation of figure 9 and application of the foregoing reasoning, a width of 50 or 70 threshold units (5 or 7 volts) appears optimum.

A certain amount of amplification drift occurs. If the window chosen is narrow, there is a possibility that, in the course of a count, the peak may move partly or entirely out of the window, thus giving an anomalously low count. If, on the other hand, the window is wide, there is a possibility that parts of adjacent peaks of components not being measured may move into the channel, thereby raising the counting rate. The choice of a window as wide as 50 or 70 units would preclude the first possibility. A test was run assuming a drift of 3.5 per cent and it was found that, on the average, the 70 unit window gave more stable results than the 50 unit

window; therefore a 70 unit width was chosen for operation.

V Drift Correction

Long and short period variation in amplification (drift) occurs despite rigid control on the voltage of the linear amplifier and the high voltage. Slight variations in temperature and humidity are primarily responsible for the short period drift. The longer period drift is not specifically assignable but is probably due to the aging of the electron tubes in the spectrometer. This drift cannot be eliminated.

Two methods of correcting for drift, both utilizing the frequent calibration of the amplification with a standard Cs^{137} source, have been developed. The first is the location of the threshold position of the Cs^{137} peak followed by calculation of the position of higher energy peaks using their known gamma-ray energies in simple proportion and a corresponding adjustment of higher energy channel positions. For example, if the 0.661 Mev Cs^{137} peak is at 210 threshold units then the thorium 2.62 Mev peak will be at 840 threshold units; if the Cs^{137} is at 215 threshold units then the thorium 2.62 Mev peak will be at 860 threshold units, etc. This method introduces a slight error due to the stretching and contraction of

the peaks which are nevertheless measured with a window of constant width. This error was checked by comparing counts made with the thorium 2.62 Mev peak at $\pm$ 3.5 per cent of its central position. Variation was found to be within statistical counting error.

The second method of correction involves movement of the Cs^{137} peak back to its standard position, 210 threshold units, by adjustment of the amplification. This would be the ideal correction were it not that the amplification may only be controlled in discrete jumps, which are too large to be of use for small scale drift.

The method actually used is a combination of these two procedures. For small-scale drift the channels are moved to the calculated position of the peaks. When the Cs^{137} peak drifts more than 6 threshold units from its standard position a 0.1 unit adjustment of the fine gain is made to move the peak approximately 10 threshold units, putting it closer to its standard position. Correction by the first method may then be made.

VI Choice of Geometry and Standards

The choice of geometry of the samples and the geometry of the standards used to calibrate these samples must be considered. The optimum sample is one from which the greatest proportion of emitted gamma-rays

impinges on the crystal. To this end, an annular cannister was designed to cover the crystal on all sides but the base with approximately 1 cm of powdered rock. Self-absorption in a source of this thickness is insignificant (Hurley, 1956; Adams et al., 1956). The weight of sample in this cannister is 600 to 700 grams.

The calibration of such a cannister was done directly for potassium by filling a cannister with a known amount of K_2CO_3 ; not enough uranium and thorium standards were available, however, to fill the cannisters; therefore the calibration had to be done indirectly with a small, 1 cm thick can of 2π geometry. The geometry is similar to that of the large cans but not exactly the same; therefore a geometry correction factor of 0.96 ± 0.03 must be applied (Adams et al., 1956). Calibration of this geometry was carried out, and several unknowns were counted.

Certain detrimental factors are present with this geometry. The indirect calibration introduces an error which is not accurately known. A sample of 600 grams is good as far as analysis is concerned but is often difficult to obtain, since the average hand specimen is considerably lighter than this. Lastly, the cannisters are expensive to make, therefore of limited number. This requires that samples be continually packed and unpacked; once a sample has been unpacked there is no way of

checking that sample again without considerable difficulty and absolutely no way of counting it again with the material in the same spacial distribution. This fact effectually precludes systematic re-running of samples for quality control.

The use of a smaller, cylindrical, 8 ounce (27 ml) can, 3.8 inches in diameter and 1.4 inches thick, eliminates many of these difficulties. Enough uranium and thorium standards were available to fill these cans and to carry out calibration directly, thus eliminating the calibration geometry factor. Less sample (350 grams) is needed to fill these cans. Lastly the cans are sufficiently available, being a stock item in many chemical supply houses, and cheap enough that they can be used as permanent containers for the samples, making systematic quality control and recounting a simple matter.

A group of standards of known uranium and thorium contents, prepared by the U.S. Atomic Energy Commission at New Brunswick, were used as the calibration standards of the present gamma-ray spectrometer. These standards were weighed on an analytical balance, and all the uranium standards and then all the thorium standards were combined. Each of these combined standards was thoroughly homogenized, packed in one of the 8 ounce cans, and then sealed. The

uranium standard, composed of uraninite diluted with dunite, weighs 354.38 grams and contains 59,827 micrograms (168.75 ppm) of uranium. The thorium standard, composed of monazite diluted with dunite, weighs 322.93 grams and contains 166,396 micrograms (515.27 ppm) of thorium and 6,575.9 micrograms (20.363 ppm) of uranium.

The potassium standard, composed of K_2CO_3 , weighs 265.4 grams, of which 150.17 grams are potassium.

The thickness of these cans and the difference in weight (and therefore in density) between these standards, and between their weights and the subsequently standardized weight of samples, 350 grams, makes it necessary to investigate the effect of absorption. Two absorption effects can introduce error in samples of different density: first, a direct attenuation of gamma-rays; and second, a difference in differential attenuation of high and low energy gamma-rays. Using absorption curves of Moteff (1955) for two densities which are roughly equivalent to the densities of the potassium and the thorium standards and considering the two extreme gamma-ray energies, the 0.61 and the 2.62 Mev, the following results are obtained:

Mev	Specific density	% gamma penetrating	% difference
0.61	1.45	78.6	3.7
0.61	1.14	81.6	
2.62	1.45	90.0	1.7
2.62	1.14	91.5	
Diff. between 0.61 and 2.62 Mev @ 1.45...11.5			
Diff. between 0.61 and 2.62 Mev @ 1.14... 9.9			
			<u>1.5</u> 15%

Differences in total attenuation for the two densities are insignificant. The difference in differential absorption is greater, about 15 per cent; however, this is not very significant because the differential absorption itself is so slight. The figures presented here are for narrow-beam absorption which is not strictly applicable to self absorption; therefore these values, small as they are, are high by unknown factors.

To test the absorption difference and slight difference in geometry of the two types of sample cans under discussion, the same sample (the Loe Shale) was counted in each of the cannisters. The resulting counts were directly proportional to the weights in each of the cans, within the 95% confidence level, indicating that differences in geometry and absorption are not significant compared to the normal operational statistical error.

One analytical drawback of this method is the small weight of the sample, which reduces the count to true

background ratio. For samples of low activity, particularly in the high energy channels, this becomes a very important source of error, which could be reduced by the use of larger samples. The activities of the granitic rocks are generally sufficiently high so that the direct calibration and quality control possible with the small cans increases the accuracy of the method more than the decrease in count to true background ratio decreases it. It is felt, however, that for samples of lower activity, having less than approximately 5 ppm thorium and 2 ppm uranium, the larger cans could be used with benefit.

VII Calculations

In the derivation of calculation equations the following terminology will be used:

R_1, R_2, R_3, R_4	Total counting rate per minute in channels 0.61, 1.47, 1.76, and 2.62 Mev with a window of 70 units (7 volts), corrected for background.
B_1, B_2, B_3, B_4	Background counting rate in channels 0.61, 1.47, 1.76, and 2.62 Mev.
T_1, T_2, T_3, T_4	Counting rates of the pure standards thorium (T), corrected for uranium contribution; uranium (U); and potassium (K) in channels 0.61, 1.47, 1.76, and 2.62 Mev respectively, all corrected for background.
U_1, U_2, U_3, U_4	
K_1, K_2	
$U^{1.76}$ cts, $Th^{2.62}$ cts, etc.....	Counts due to uranium, thorium etc. alone; i.e., that component of the total count in channel 1.76, 2.62, etc. Mev. Capitalized letter indicates component; superscript figure indicates channel.

$U^{1.76}$, $Th^{2.62}$, etc. Total uranium or thorium in
ppm; potassium in per cent
calculated from channel indicated
in the superscript.

W Total weight of sample; in most
cases this is 350 grams.

The counting rates in the various channels due to true background (no sample in the detector) are as follows: $B_1 = 33$ cts./min.; $B_2 = 8$ cts./min.; $B_3 = 2$ cts./min.; $B_4 = 1$ ct./min. These counting rates are constant within statistical limits and have been counted over a period of several months.

The standards were counted numerous times over a period of a week. An average figure was obtained for each channel, and average background in that channel was subtracted. The uranium has no thorium component, and therefore the uranium contribution to the thorium standard counts could be calculated easily; since the counting rate is known for 59,827 micrograms of uranium in the uranium standard, a simple proportion determines the expected contribution from 6,575 micrograms of uranium in the thorium standard. This expected amount was then subtracted from the total counting rate of the thorium standard to give the effect of pure thorium. The counting rates in counts/min. of pure standards are as follows:

$T_1 = 1372$	$U_1 = 1246$	$K_1 = 242$
$T_2 = 130.5$	$U_2 = 224.8$	$K_2 = 646$
$T_3 = 101.3$	$U_3 = 219.4$	$K_3 = 0$
$T_4 = 169.6$	$U_4 = 3.21$	

A series of ratios were calculated with these figures in order to strip out contribution of counts in lower channels based on known pure counting rates in higher energy channels. These calculations were predicated on the fact that the relative heights of the peaks of a pure spectrum are constant. Unfortunately, uranium contributes a small number of counts to the high 2.62 thorium channel. The counts due to thorium alone in this channel may be calculated as follows: Let

x = counting rate in R_4 due to thorium alone (Th 2.62)
then x (cts)

$x (T_3/T_4) =$ counting rate in R_3 due to thorium alone.

$(R_4 - x) =$ counting rate in R_4 due to uranium alone.

$(R_4 - x) U_3/U_4 =$ counting rate in R_3 due to uranium alone.

and

$$R_3 = (R_4 - x) U_3/U_4 + x(T_3/T_4)$$

$$R_4(U_3/U_4) - x(U_3/U_4) + x(T_3/T_4)$$

$$x(T_3/T_4 - U_3/U_4) + R_4 (U_3/U_4)$$

or

$$x = \frac{R_3 - R_4 (U_3/U_4)}{(T_3/T_4 - U_3/U_4)} = (R_4(U_3/U_4) - R_3) \left(\frac{1}{U_3/U_4 - T_3/T_4} \right)$$

$$= R_4 \left(\frac{U_3/U_4}{U_3/U_4 - T_3/T_4} \right) - R_3 \left(\frac{1}{U_3/U_4 - T_3/T_4} \right)$$

All the figures in the parentheses are known counting rates; by substituting the above values, this equation becomes

$$(1) \quad \text{Th}_{\text{cts}}^{2.62} = R_4 (1.0088) - R_3 (0.01473)$$

By using the ratio T_3/T_4 , $\text{Th}_{\text{cts}}^{1.76}$ may be calculated.

$$\text{Th}_{\text{cts}}^{1.76} = \text{Th}_{\text{cts}}^{2.62} (T_3/T_4)$$

then

$$\text{U}_{\text{cts}}^{1.76} = R_3 - \text{Th}_{\text{cts}}^{2.62} (T_3/T_4)$$

By substitution of standard counting rates this expression becomes

$$(2) \quad \text{U}_{\text{cts}}^{1.76} = R_3 - \text{Th}_{\text{cts}}^{2.62} (0.5973)$$

The potassium contribution to R_2 may be calculated by stripping off counts due to both uranium and thorium in this channel calculated from their counting rates at higher energy levels.

$$\text{K}_{\text{cts}}^{1.47} = R_2 - \text{Th}_{\text{cts}}^{2.62} (T_2/T_4) - \text{U}_{\text{cts}}^{1.76} (U_2/U_3)$$

this becomes

$$(3) \quad \text{K}_{\text{cts}}^{1.47} = R_2 - \text{Th}_{\text{cts}}^{2.62} (0.7694) - \text{U}_{\text{cts}}^{1.76} (1.0255)$$

The fourth channel may be used to check any of the others. It is most efficient for a check of uranium because it is centered on the 0.61 Mev uranium peak. If the calculation agrees with the uranium determined from the 1.76 Mev channel then all the counts, uranium, thorium

and potassium must be correct since an error in counting rate or in calculation at any point would result in a discrepancy in the two uranium figures. Calculation of thorium in this channel does the same thing; when done in conjunction with a uranium check, however, it checks only the arithmetic calculation of $\text{Th}^{2.62}$. The equations for this determination are:

$$\text{U}_{\text{cts}}^{0.61} = R_1 - K_{\text{cts}}^{1.47} (K_1/K_2) - \text{Th}_{\text{cts}}^{2.62} (T_1/T_4)$$

and

$$\text{Th}_{\text{cts}}^{0.61} = R_1 - K_{\text{cts}}^{1.47} (K_1/K_2) - \text{U}_{\text{cts}}^{1.76} (U_1/U_3)$$

which become

$$(4) \quad \text{U}_{\text{cts}}^{0.61} = R_1 - K_{\text{cts}}^{1.47} (0.3746) - \text{Th}_{\text{cts}}^{2.62} (8.0896)$$

and

$$(5) \quad \text{Th}_{\text{cts}}^{0.62} = R_1 - K_{\text{cts}}^{1.47} (0.3746) - \text{U}_{\text{cts}}^{1.75} (5.6843)$$

These equations solve for unknowns in terms of counting rates; these counting rates must be calibrated in absolute terms of ppm thorium and uranium and per cent potassium. In the following discussion U micrograms of uranium in standard, T micrograms of thorium in standard, and K centigrams of potassium in standard; T_4 , U_3 etc. as before. Then the proportion may be set up

$$T/T_4 = W/\text{T}_{\text{cts}}^{2.52}$$

But, since W is in grams and T in micrograms, this is equivalent to

$$\text{ppm Th}^{2.62} = \frac{(T/T_4) T_{\text{cts}}^{2.62}}{W}$$

but

$$T/T_4 = \frac{166.396.32}{169.5} \quad 981.11$$

therefore

$$\text{ppm Th}^{2.62} = \frac{981.11}{W} \times T_{\text{cts}}^{2.62}$$

if W = 350 grams

$$(6) \quad \text{ppm Th}^{2.62} = 2.803 (T_{\text{cts}}^{2.62})$$

By an exactly analogous calculation the calibration equation for uranium is

$$(7) \quad \text{ppm U}^{1.76} = 0.7798 (U_{\text{cts}}^{1.76})$$

The potassium content of the standard is given in centigrams, therefore, by using the same type of proportion, the final calibration is in per cent.

$$(8) \quad \text{per cent K}^{1.47} = 0.06642 (K_{\text{cts}}^{1.47})$$

The expressions for the 0.61 Mev channel are then

$$(9) \quad \text{ppm U}^{0.61} = 0.137 (U_{\text{cts}}^{0.61}) \quad \text{and}$$

$$(10) \quad \text{ppm Th}^{0.61} = 0.3465 (Th_{\text{cts}}^{0.61})$$

Combining the stripping equations (1-5) with these calibration equations (5-10), the following equations are obtained. These are the equations used in actual calculation:

$$\text{Th}^{2.62} = 2.803 \left[R_4(1.009) - R_3(0.0147) \right]$$

$$\text{U}^{1.76} = 0.78 \left[R_3 - (\text{Th}_{\text{cts}}^{2.62})(0.5973) \right]$$

$$\text{K}^{1.47} = 0.0664 \left[R_2 - (\text{U}_{\text{cts}}^{1.76})(1.0255) - (\text{Th}_{\text{cts}}^{2.62})(0.7694) \right]$$

$$\text{U}^{0.61} = 0.137 \left[R_1 - (\text{K}_{\text{cts}}^{1.47})(0.3746) - (\text{Th}_{\text{cts}}^{2.62})(8.0896) \right]$$

$$\text{Th}^{0.61} = 0.3465 \left[R_1 - (\text{K}_{\text{cts}}^{1.47})(0.3746) - (\text{U}_{\text{cts}}^{1.76})(5.6843) \right]$$

VIII Preparation of Samples

Depending on the grain size of the sample, 700 to 1000 grams were broken on a hydraulic rock crusher and with a geological hammer to approximately 1/4 inch size. This material was run through a rotary-type pulverizer which reduced all the material to -60 mesh and most of it to -100 mesh. A few of the first samples to be ground were thoroughly homogenized on a rotary blender following this crushing, and 350 grams of this powder was packed into an 8 ounce can and sealed. The later samples were ground completely to -100 mesh; the final grinding of the -100 mesh material was done with a mullite mortar and

pestle. The powder was then homogenized, packed, and sealed. All samples were allowed to re-attain radioactive equilibrium with respect to radon, which might have been lost in the pulverizing process, by keeping the powder in the cans for at least one week prior to counting.

All apparatus was thoroughly cleaned between each crushing, and precautions were taken against dusting loss. Greater than 99 per cent recovery was attained; the loss was mostly in the coarse breakage.

The pulverizing process was checked for contamination by grinding a very hard sample of known activity (Em-1) and then counting it. There was no perceptible increase in activity. Separate 1000 gram lots of a single sample, the Vermillion granite, were crushed by separate operators five months apart; the results were within statistical error.

IX Error, Accuracy, and Length of Count

Precision: The precision of the present instrument is a function of many factors. The major source of error, up to a certain number of counts, is statistical counting error. Other error - such as true variation in background; drift of photo-peaks due to amplification variation; slight variation in sensitivity of the instrument; and error, however slight, in drift correction and setting

of the instrument - are physical errors and cannot be quantified without extensive and probably unprofitable investigation. For fairly short counts the statistical counting estimate of error was felt to be roughly correct, but with increasing count length the physical error factors become more significant.

The statistical error is a function of the total number of counts and of total background count. The standard deviation of the total count is very well approximated by the square root of the total number of counts; the standard deviation of the background count is the square root of the background count. The total number of counts from the sample is obtained by subtracting the background count from the total count, and the error associated with the sample count is found by compounding the error in standard statistical fashion, i.e. the square root of the sum of the squares of the two standard deviations. This problem has been dealt with by Loevinger and Berman (1951). They have drawn up a series of curves which define the relation between these two variables and the per cent standard deviation for the sample count.

In this analytical method the total sample count is not important; it is the portion of the total count due to only one component (thorium, uranium, or potassium) which is of interest. In any given channel the contribution

from the other components is a function of the ratio of their abundances. This may be determined, and the contribution may be calculated and considered as background. The total to (compound) background ratio will then be known, and the Loevinger and Berman chart may be used to obtain per cent standard deviation directly.

By consideration of a rock with an average amount of thorium, uranium, and potassium it is possible to specify the number of counts necessary in various channels to obtain a given per cent standard deviation. If the average granitic rock is considered to have 12 ppm thorium, 3.5 ppm uranium, and 4.5 per cent potassium, then the counting rates in channels 2.62, 1.76, 1.47, and 0.61 Mev will be approximately 6, 10, 88, 130 counts per minute respectively. For such a rock, 5 per cent standard deviation may be attained by counting 1000 counts in channel 2.62, 6000 counts in channel 1.76, and 1000 counts in channel 1.47. These counts will take 3, 10, and $\frac{1}{2}$ hours each in their respective channels. With counts of this length it was felt that the statistical error should be a fairly good approximation of the true reproducibility. This contention has been born out by subsequent observations.

Packed samples were sequentially numbered by can regardless of the sample collection number. A system of quality control based on these numbers was initiated in

order to check the stability of the instrument and to determine the empirical reproducibility (precision) of the method. Each tenth sample which was counted and whose number was a multiple of 10 was immediately succeeded by a previously counted sample whose can number was a multiple of 5. Such cans were re-run in order of increasing number. In this way six samples were counted twice. During part of this investigation a single sample was counted three separate times immediately following each tenth sample whose can number was a multiple of 10.

Standard deviations have been calculated for each of these 7 samples for thorium, uranium, and Th/U ratio; and their averages have been calculated (see table 12). The number of samples is too small to justify putting much weight on these data; it is significant, however, that the statistical estimates are in approximate agreement with the observed deviation. The uranium error is greater than the thorium error, as is expected on several grounds: first, the count length in the 1.76 position is very much longer, and the physical error probably plays a larger part than in the other channels; second, the thorium contribution to the 1.76 channel is large and must be considered as a constant for calculation of count to (compound) background ratio (however, there is a statistical error associated with this figure which is ignored); and

third, the "topography" of the spectrum is much more irregular at the 1.76 channel than at the 2.62 channel (see figure 9), and therefore the drift affects the lower energy channel more.

No. of Repeti- tions	Sample	Can#	Observed % Std. Dev.		% Std. Dev. From Counting Statistics	
			Th	U	Th	U
2	Sc 12-60	20	0.93	8.10	3.90	3.75
2	JW 32	24	2.92	9.30	3.75	2.90
2	E 5-404	46	3.69	2.47	4.80	4.80
2	E 4-403	44	2.82	6.80	3.55	4.65
2	Sc 25-72	41	5.90	1.01	5.10	5.75
2	E 10-15	26	3.34	0.90	3.80	4.70
3	Sc 1-49	19	4.70	9.75	4.70	3.50
Ave.			3.47	5.48	4.23	4.29

Table 12. - Comparison of estimated relative analytical error (precision) calculated by using statistical considerations, compared with observed reproducibility.

The precision of the Th/U ratio is a function of the errors associated with the determination of thorium and uranium. The expression for the error of a ratio of errors is a standard statistical combination (Friedlander and Kennedy, 1956, p. 262). If 5 per cent standard deviation is associated with each of the thorium and uranium determinations, their ratio will have a standard deviation of 7.1 per cent. Reduction of the standard deviation of either the uranium or the thorium to 3 per cent, by increasing the number of counts by a factor of two, decreases the error of the ratio only 1.3 per cent. Thus it is difficult to significantly improve the precision of the ratio without increasing the counting periods rather drastically.

Accuracy: The accuracy of the method depends upon the factors affecting the precision plus some additional factors. The initial calibration is entirely dependent on the accuracy of the U.S. Atomic Energy Commission standards and on the re-weighing and mixing of these standards at The Rice Institute. The counting rates of the standards are considered constant in all calculations. This is not entirely valid because there is a small statistical uncertainty; however, the mathematical complexity of carrying the statistical errors through the

derivation of calculation-equations is so excessive as to make it impractical. The accuracy cannot be less than ± 5 per cent but is probably less than ± 15 per cent.

The interlaboratory and intralaboratory reproducibility of determinations is presented in table 13. Several operational sedimentary rock standards have been established and have been checked by Shell Development Company using gamma-ray spectrometry; by the U.S. Geological Survey with wet chemical methods (personal communication); and by The Rice Institute using gamma-ray spectrometry, checked by fluorimetric uranium and wet thorium determinations. The standard rock samples G-1 and W-1 have been analyzed by Smales (1956) and also by gamma-ray spectrometry; these are included in table 13. Several granitic rocks involved in the present investigation have been checked by Shell Development Company (Adams et al., 1957) and are also included in the table. Most of these analyses have Rice chemical checks as well.

The interlaboratory reproducibility of analyses is the best estimate of accuracy. The agreement of the data in figure 3 constitutes strong evidence for the validity of the initial calibration of the gamma-ray spectrometer. In almost all cases the range of values exceeds any one precision estimate; this is to be expected, however, since these are only estimates of reproducibility. The values,

Table 13.- Interlaboratory and intralaboratory reproducibility of uranium, thorium, and potassium determinations.

SAMPLE	DETERMINATION	Th ppm	U ppm	K%	Th/U
Chattanooga Shale	Shell	5.2,	80,	2.7,	0.7,
	Rice	8.7	79	2.8	0.08
	U.S.G.S. Rice (Chem)	8.2, 8.2	74, 69	2.2	0.10 0.11
Lawrence Red Shale	Shell	13	1.6	2.9	8.1
	Rice	11±.5	1.8±.09	2.6±.1	6±.4
	U.S.G.S. Rice (Chem)	11.7, 10.6	3.0, 2.7 1.6		4
Heebner Shale	Shell	9.8	32	2.2	0.31
	Rice	16±.56	31±.49	2.4±.13	0.52±.03
	U.S.G.S. Rice (Chem)	8.8, 8.9	34.5, 34 27		
Lee Sandy Shale	Shell	14	3.7	2.5	3.8
	Rice	12±.6	4.2±.15	2.3±.08	2.8±.17
	U.S.G.S. Rice (Chem)	14, 13.4	5.2, 4.6 2.1		
Arkansas Bauxite	Shell	96	6.4	0.3	15
	Rice	84	4.6	0.05	18
	U.S.G.S. Rice (Chem)	79.4, 79.8	7.0, 6.6 6.4		11
Surinam Bauxite	Shell	132	6.1	0.3	21.6
	Rice	122±1.2	5.1±.5	minus no.	24±2.4
	Rice (Chem)	130	6.0		

Table 13.- (Continued).

SAMPLE	DETERMINATION	Th ppm	U ppm	K%	Th/U
Lawrence Underclay	Shell Rice (Chem)	13	4.2	2.4	3.1
		13	4.1	2.2	3.3
			2.4		
Lawrence Shale (Shale beneath Underclay)	Shell Rice (Chem)	13	4.0	2.0	3.3
		12	4.5	1.8	2.5
			3.8		
Galveston SS	Shell Rice U.S.G.S. Rice (Chem)	2.0±.1	0.5±.03	1.0±.1	3.8±.3
		2	0.6	0.66	3
		1.7	0.8, 0.8		2
			0.4		
Diabase W-1	Smales Rice	1.5	0.517	0.6	2
			0.8		
Granite STND G-1	Smales Rice	50	3.41	4.6	11.9
			4.2		
Pikes Peak Granite F 19-24	Shell Rice (Chem)	22.2	0.89	4.5	24.9
			0.85		
Pikes Peak ¹ Granite F 10-15	Shell Rice (Chem)	9.93±.39	16.93, 11.32	4.63±.18	0.88±.05
		35.4±1.5	4.65±.2	4.52±.15	7.6±.47
			2.6, 3.1		
Idaho Springs ¹ Fin F 2-7	Shell Rice (Chem)	24.81±1.52	1.61, 1.88	4.62±.18	13.22±.56
		9.5±.32	5.2±.12	4.3	1.83
		9.3	2.7, 3.3		

Table 13.- (Continued).

SAMPLE	DETERMINATION	Th ppm	U ppm	K ^o /o	Th/U
Conway Granite NH 5-307	Shell Rice	93.58±3.3 82.5±2	20.49±.8 18.6±.5	4.27±.2 4.14±.24	5.47±.24 4.4±.17
Silver Plume Granite F 32-37	Shell Rice	41.55±1.49 36.9±1.5	4.55±.6 4.6±.23	4.88±.19 4.5±.19	8.41±.56 8±.5
Spavinaw Granite Sp 2-4	Shell Rice	18.6±.6 16.8±.79	5.1±.2 4.9±.15	3.1±.1 2.9±.13	3.6±.2 3.42±.19
Killarbey Granite K 17-255	Shell Rice ^a Rice Rice (Chem)	16.45±.61 16.22 15.39±.4 14.4	2.82, 3.44 1.9 2.5±.07	4.46±.16 4.64 4.3±.1	5.83±.4 8.5 3.55±.13
Mt. Morrison Gneiss JW-32	Shell Rice	10.57±.47 8.64	4.06±.17 3.84	1.24±.07 1.25	2.6±.16 2.24

¹ Shell and my determination differ widely.² Done with original calibration and large annular cannisters. Approximate at best.

ignoring a few which are obviously discrepant, are generally within about 20 per cent.

Considering the estimated accuracy and the observed variation between laboratories, it seems unnecessary to strive for precision better than 5 per cent. Better precision than this would not aid in most geological interpretation, but the large number of samples which can be counted at this relatively low precision can provide important information.

COMPARISON WITH OTHER GAMMA-RAY SPECTROMETERS

Hurley (1956) has described the application of gamma-ray spectrometry to ore assay and to the determination of the uranium, thorium, and potassium contents of common rocks. His method is similar to the one used here. The major differences between Hurley's method and the method described in this paper are the size of the crystal and the gamma energies of the peaks counted. Hurley's crystal is 1 3/4 inches in diameter by 2 inches in height as compared with 3 inches by 3 inches of the present crystal. With his small crystal it is not practicable to measure the high energy gamma-rays because of the poor resolution and the very high Compton scatter (see figures 2 and 1C). His two low energy channels, for determination of uranium and thorium, are set at 168 and 226 kev; the measurement

is made in these channels with a 25 kev window. The potassium is measured at 1.47 Mev with a 134 kev window. The equations to solve for the unknown counting rates are more complicated since all three components are present in all channels. Due to the narrow windows, small drift has considerable effect on the counting rate; therefore Hurley must consider drift in his estimate of error. The counting rates are considerably higher in the lower ranges of gamma-energy, but counts from the unknown component being measured are so low compared with Compton spread and other background that counting is very inefficient.

The method reported from Shell Development Company by Adams et al. (1956) is essentially identical to the present method, differing only in detail. Shell utilizes a 3 by 3 1/2 inch crystal as compared with the 3 by 3 inch crystal used in this investigation. The major difference is source geometry; Shell uses a large annular canister holding approximately 600 grams of sample compared with the small (350 gram) cylindrical cans used in this investigation. The pros and cons of these containers have already been discussed.

PRESENTATION OF DATA

A total of 49 samples representing 38 bodies of granitic rock has been analysed as described previously for uranium, thorium and potassium by gamma-ray scintillation spectrometry. The results are summarized in table 14. Thin sections of these samples have been studied in detail, and modal analyses have been obtained by counting 1000 to 1500 points on one or two thin sections of each rock, depending on the grain size. These modal analyses are presented in table 15. The location and description of each of the rock bodies from which the samples were taken and descriptions of the samples are presented in the appendix.

An attempt was made to sample various different types of plutons in respect to the following criteria: 1) absolute age; 2) age relative to orogeny (pre-, syn-, and post-orogenic; 3) geographic distribution in the United States and central Canada; 4) areal relation to zone of major tectonism; 5) concordance or discordance; 6) conformability or disconformability; 7) harmony or disharmony (with respect to regional rank of metamorphism as used by Walton (1955)); 8) size of granitic body; 9) mode of genesis (differentiation, rheomorphism, or granitization) and 10) whether the body is an isolated pluton or a member of a comagmatic series.

Table 14.- Thorium and uranium contents of all granitic rocks analyzed in this investigation, arranged in order of increasing Th/U ratio.

SAMPLE	Th/U	Th _γ ppm	U _γ ppm	K _γ %	U _f ppm	Th _{chem} ppm
KA 80-408	0.2	0.07	0.3	0.8		
D 17-238	1.0	0.5	0.5	0.4		
W 13-194	1.4±.12	2.8	2.0 ±.07	1.9 ±.07		
M 3-205	1.5±.13	2.5 ±.2	1.6 ±.06	2 ±.08		
JW-70	1.6±.15	3.1 ±.25	1.9 ±.08	1.8 ±.09		
S 14-166	1.8±.2	4.6 ±.32	2.6 ±.22	1.4 ±.05		
P 2-7	1.8±.14	9.2 ±.5	4.6 ±.1	4.3 ±.1	3.0	9.3
NH 47-349	2.0±.11	11 ±.6	5.5 ±.12	3.0 ±.12		
Y 24-119	2 ±.1	16 ±.4	8.3 ±.2	2.51 ±.12		
Y 1-96	2.2±.13	8.2 ±.2	3.8 ±.13	2.1 ±.09		
P 11-141	2.2±.18	3.4 ±.33	1.5 ±.08	1.5 ±.05	1.2	0.6
JW-32	2.3±.11	8.8 ±.35	3.8 ±.11	1.3 ±.1		
D 5-226	2.5±.15	8.7 ±.5	3.5 ±.15	2.0 ±.07		
KA 6-405	2.5±.13	12 ±.5	4.7 ±.14	3.5 ±.14		
M-4	2.6±.13	12 ±.5	4.6 ±.14	2.4 ±.08		
P 15-145	2.7±.2	4.5 ±.3	1.7 ±.1	1.5 ±.06		
D 9-230	3.1±.2	8.4 ±.5	2.8 ±.12	1.8 ±.21		
CB 3-45	3.1±.15	12 ±.5	4.0 ±.14	3.2 ±.1		
So 1-49	3.2±.2	10 ±.2	3.2 ±.1	3.3 ±.2		
So 15-63	3.2±.36	3.2 ±.25	1.0 ±.08	0.4 ±.04		
W 3-399	3.2±.16	12 ±.5	3.8 ±.11	3.4 ±.08		
A 13-268	3.3±.24	7.4 ±.44	2.3 ±.09	6.2 ±.15		
So 12-60	3.3±.18	11 ±.5	3.5 ±.13	2.3 ±.04		
P 13-143	3.3±.23	5.4 ±.25	1.7 ±.08	1.5 ±.06		
M 6-208	3.4±.34	3.5 ±.28	1.1 ±.06	1.6 ±.07		
A 4-259	3.4±.25	5.0 ±.8	1.5 ±.07	4.5 ±.17	3.1	
Sp 2-4	3.4±.18	17.0 ±.5	4.9 ±.1	2.9 ±.13		
So 25-72	3.5±.2	9.7 ±.6	2.8 ±.14	2.1 ±.1		
MW 3-76	3.5±.19	15 ±.4	4.5 ±.16	2.7 ±.07		
Y 7-102	3.6±.24	8.3 ±.35	2.3 ±.1	2.4 ±.1	1.73	14.1
NH 43-345	3.6±.19	14 ±.35	3.8 ±.17	3.0 ±.08		

Table 14.- (Continued).

SAMPLE	Tn/U	Th _γ ppm	U _γ ppm	K _γ %	U _f ppm	Th _{chem} ppm
Y 9-104	3.7±.22	13	3.5 ±.13	1.2 ±.08		
So 8-56	3.9±.23	11	3.0 ±.12	2.5 ±.1		
NH 29-331	3.9±.17	22	5.5 ±.18	3.5 ±.1		
F 16-21	4.3±.21	22	5.0 ±.23	3.2 ±.1	3.4	
D 14-235	5.2±.35	13	2.5 ±.11	2.7 ±.09		
P210G(407)	5.3	0.75	0.14	0.88		
E 4-403	5.5±.3	21	3.9 ±.16	4.1 ±.14		
E 3-402	5.7±.6	22	3.8 ±.04	4.2 ±.12		
E 5-404	6.0±.3	12	2.0 ±.08	3.4 ±.1		14.4
K 17-255	6.7±.13	15	2.5 ±.07	4.3 ±.1		
F 32-37	8.0±.5	37	4.6 ±.23	4.5 ±.19		
12M	8.3±.1	5.2	0.63±.06	2.8 ±.13		
KA 72-406	8.3	1.1	0.13	1.0		
KA 27-410	8.8±.7	18	2.0 ±.14	2.5 ±.12		
Id 7-202	9.2±.92	7.7	0.84±.07	3.1 ±.12		
V 1-409	9.6±.4	30	3.1 ±.14	4.2 ±.16		
F 10-15	10 ±.5	39	3.9 ±.16	4.5 ±.15	3.0	
F 19-24	25 ±6.8	22	0.89±.15	4.5 ±.2	0.85	
Average	4.42	11.35	2.96	2.69		

¹ Fluorimetric uranium determination.

Table 15.- Modal analyses of granitic rocks studied, arranged in order of increasing R.

SPECIMEN	R'	C.I. ²	QUARTZ	PLAGIO- CLASE ³	POTASH FELDSPAR	BIOTITE	HORNBLende	OPaques	ZIRCON	APATITE	SPHENE	EPIDOTE	ALLANITE	MUSCOVITE	CHLORITE
W 3-399		5.7	30	79 ²⁸	64	1.03	2.6	2.06	Tr	Tr	Tr	Tr			
K 80-(408)		3	18	41 ³⁰		3.0			Tr- ⁴	Tr	Tr	Tr			
Y 9-104	0.01	3.3	56	81 ¹⁸	0.22	3.3			Tr	Tr	Tr+ ⁵	Tr+			Tr
P210G(407)	0.01	9.0	14	40 ⁵⁰	0.75	1.35	7.5	Tr	Tr	Tr	0.03	0.1	0.01		Tr
So 15-63	0.02	42.3	12	55 ⁴¹	0.8	21.1	47.3	Tr	Tr	Tr	Tr+	Tr	?		Tr+
W 13-194	0.03	24	19	58 ³⁰	1.5	2.0	2.9	Tr	Tr	Tr	Tr+	4.0		2.2	Tr
KA 72	0.03	8.2	32	62 ⁴⁸	1.9	6.2	7.2	0.5	Tr	Tr	Tr	Tr			Tr
S 14-166	0.06	14	20	55 ⁴⁰	3.9	9.2	2.8	Tr	Tr	Tr	Tr	Tr			Tr
P 15-145	0.06	12	30	49 ⁴⁰	3.3	14	6.0	0.1	Tr	Tr	Tr	Tr		Tr	
P 11-141	0.07	20	24	53 ⁴⁰	4.0	7.8	6.6	0.35	Tr	Tr	Tr+	0.1		0.18	
P 13-143	0.07	15	33	43 ³⁸	3.3	10	4.2	0.2	Tr	Tr	Tr	0.1			
JW-70	0.12	14	37	56 ³⁷	5.3	7.1	6.3				0.2				Tr
Y 1-96	0.14	14	23	54 ³¹	7.8	8.2	0.03			0.2	0.03				
M 4-A	0.19	9.1	27	52 ³⁰	10	4.2	2.4	0.3			0.6				
M 3-205	0.2	7.7	25	59 ³⁸	11	7.2									
Y 7-102	0.21	8.1	28	41 ²⁸	11	5.0	12	1.7		Tr				0.15	Tr
M 6-208	0.25	20	12	48 ³⁷	10	8.1	1.3	0.2	Tr	Tr	0.5	0.65			Tr
So 8-56	0.32	9.6	36	46 ²⁰	13	6.9	0.9	0.03	Tr-	Tr	0.03	0.02	Tr		
Id 7-202	0.32	7.0	30	39 ³⁸	15	3.4	Tr	0.3	Tr	Tr	Tr				Tr
So 12-60	0.33	4.6	35	49 ¹⁸	15	3.9		Tr	Tr	Tr	Tr				
So 25-72	0.35	3.9	44	35 ²⁰	13	1.7		0.1	Tr	Tr	Tr				
12M	0.36	1.8	31	38 ²⁷	18	9.5		0.59	0.02	Tr	0.06				
E 3-402	0.53	8.6	26	31 ¹⁸	28	10.8			Tr	Tr	Tr		0.03		
E 5-404	0.58	11	29	42 ¹⁸	22	2.4			Tr	Tr	Tr		?		
KA 27-410	0.58	2.4	31	31 ¹⁸	24	2.5			Tr	Tr	Tr			Tr	
NH 47-349	0.66	7.1	40	31 ¹⁸	20	2.5			Tr	Tr	Tr				

Table 15.-- (Continued).

SPECIMEN	R ¹	C.I. ²	QUARTZ	PLAGIO- CLASE ³	POTASH	FELDSPAR	BIOTITE	HORNBLende	OPaques	ZIRCON	APATITE	SPHENE	EPIDOTE	ALLANITE	MUSCOVITE	CHLORITE
E 4-403	0.68	10	44	26 ₂₀	18		10	0.01	0.08	Tr	Tr	Tr+		0.01		
So 1-49	0.73	2.9	40	33 ₁₈	24		2.9		Tr	Tr+						
F 16-21	0.78	14	28	33 ₂₀	25		11		1	Tr	1	Tr		Tr+		
KA 6-405	0.8	14	18	38 ₂₈	30		9.6	4.6	Tr	Tr	Tr	Tr	Tr+			
NH 29-331	0.82	2.3	37	33 ₁₈	27		2		0.2	Tr	Tr	Tr				
NH 43-345	1	15	52	17 ₂₇	17		12		0.3	Tr	Tr		Tr			
F 10-15	1	3.4	31	32 ₂₀	33		2.8			Tr	Tr		Tr			
V 1-409 (a)	1.07	3.1	24	35 ₁₈	38		1.5			Tr			Tr		0.9	Tr
V 1-409 (b)	1.25	3.6	27	30 ₁₈	38		3.6		0.01	Tr	Tr		Tr	Tr+	0.1	Tr
F 32-37 (a)	1.43	4.5	32	26 ₂₀	38		2.8		0.2	Tr	Tr		Tr	Tr+	2.2	Tr
F 32-37 (b)			38	30 ₂₀	27		2.9		Tr	Tr+	Tr+			Tr	Tr	
F 19-24 (a)	1.6	4.7	19	31 ₃₁	45		4.1		1.3	Tr	Tr+					
F 19-24 (b)			38	21 ₃₁	35		3.4			Tr						
A 4-259	1.6	17	28	22 ₂₀	35			14		Tr	Tr					
F 2-7	1.8	8.6	33	21 ₁₀	36				0.7	Tr	Tr	Tr			7.9	
K 17-255 (a)	1.7	3.8	23	26 ₁₂	45		3.2		0.6	Tr	Tr					
K 17-255 (b)	3.1	3.3	32	15 ₁₂	50		3.3		Tr	Tr	Tr					
A 13-268	4.8	7.1	20	12	60		5.3		2.1	Tr	Tr	Tr				Tr

¹ R is potassium feldspar/ plagioclase ratio.² C.I. is color index.³ Subscript indicates per cent anorthite.⁴ Tr- is very scarce trace mineral.⁵ Tr+ is abundant trace mineral.

The collection is primarily intended to be used in analysis of large scale variations of uranium and thorium; therefore, in the group of analyses presented here, no extensive study has been made of sampling variation nor of within-pluton variation. Several preliminary tests have been run, however, in order to obtain some independent information on these two levels of variation which could obscure larger scale relationships.

Sampling homogeneity (hand specimen level) has been tested on several samples. A large sample of medium-, to coarse-grained Pikes Peak granite (F 19-24), in which irregular distribution of uranium might be expected because of the large grain size, was broken into 6 pieces ranging in weight from 30 to 432 grams. These pieces were crushed separately and analysed for uranium by the fluorimetric method. Results for all the sub-samples are within the analytical error of the method, which is estimated to be 15 per cent. A sample of the medium-grained Spavinaw granite (Sp 2-4) was divided into three sub-samples which were separately crushed and analyzed; the results are within analytical error. Two 1000 gram portions of the medium-, to coarse-grained Vermillion granite (V 1-409) were separately pulverized and analyzed by gamma-ray scintillation; the results are

within analytical error (approximately 5 per cent). These data tend to show that sampling variation is not significant.

Data from the literature has demonstrated that the thorium and uranium contents of rocks commonly vary systematically within individual plutons. Several groups of samples from among those analyzed in the course of this study are composed of replicate samples taken from the same plutons; therefore, some preliminary data are available on variation of uranium and thorium within individual plutons.

Three samples, P 11-141, P 13-143, and P 15-145, were collected two miles apart, from the contact toward the central portion of Bucks granodiorite in the northern part of the Sierra Nevada. From table 15 it may be seen that the mineralogy of these samples is almost the same; the major differences are in the total amount of mafic material and in the biotite-to-hornblende ratio. In these samples the thorium content varies from 3.4 to 5.4, the uranium content varies from 1.5 to 1.7, and the Th/U ratio varies from 2.2 to 3.3. These variations are very small and not far from analytical error for samples of such low activity.

The Enchanted Rock batholith of Texas is represented by three samples collected in the central portion of the

batholith within 200 feet of one another. Two of the samples (E 3-402 and E 4-403) are from the petrographically homogeneous porphyritic facies. The uranium and thorium contents of these two samples are almost exactly the same (see table 14). The third sample (E 5-404) is from an aplite dike which cuts the porphyritic facies. It is mineralogically very similar to the coarse facies but entirely different texturally. The thorium and uranium contents of this aplite are both about one half that of the porphyritic rock, and therefore, the Th/U ratio is almost the same as that of the porphyritic facies.

Two samples of the Cathedral Peak granite were collected from the central portion of the Sierra Nevada. One sample (MW 3-76) is from the Mount Whitney area and the other (Y 24-119) is from Yosemite National Park, 100 miles north of Mount Whitney. The rocks are very coarsely porphyritic, and therefore, no attempt was made to point count them. The samples, however, are very similar to one another both megascopically and microscopically. In both rocks thorium is high and of about equal abundance; uranium, however, is high in sample Y 24-119 (8.3 ppm) but only slightly above average in sample MW 3-76 (4.5 ppm). This difference in uranium content results in Th/U ratios of 2 and 3.5, respectively.

Three samples from the Idaho batholith have been examined. Sample 12M is from the light inner facies of the batholith, has a low content of thorium (5 ppm) and uranium (0.6 ppm), and has a Th/U ratio of 8.1. Samples M-4 and Id 7-202 were collected approximately 30 miles apart in the mafic outer facies of the batholith and are texturally and mineralogically distinct from 12M (see table 15). These two samples are petrographically very similar to one another, but they differ widely in thorium and uranium contents. Sample 4M contains approximately average amounts of uranium and thorium and has a Th/U ratio of 2.6. Sample Id 7-202 is low in thorium (7.7 ppm), extremely low in uranium (.8 ppm), and has a Th/U ratio of 9.2.

Two samples from the Pikes Peak batholith of the Colorado Front Range have been studied and analyzed. Sample F 10-24 is from within 300 feet of the contact of the Pikes Peak granite with the Boulder Creek granite which it intrudes. Sample F 10-15 is from another part of the batholith, one-half mile from the contact. The two rocks are similar petrographically, but plagioclase composition, the potash-feldspar/plagioclase ratio, and the color indices differ slightly. In both rocks the thorium content is extremely high. In sample F 19-24 the uranium is very low, resulting in a Th/U ratio of 25,

the highest found in the present study. Sample F 10-15 contains roughly an average amount of uranium but has a high Th/U ratio of 10, due to the extremely high thorium content.

Two samples from the Marysville stock have been examined. Sample M 3-205 is from the center of the stock, and sample M 6-208 is from the contact of the stock with the Helena dolomite. The petrography of the two samples is similar, but M 6-208 has three times as much hornblende and only one half as much quartz as sample M 3-205. The desilication may be a result of contact relations with the dolomite. The thorium content increases from the center to the border of the stock from 2.5 to 3.5 ppm; the uranium, however, decreases from 1.6 to 1.1 ppm. Thus the Th/U ratio increases from 1.5 to 3.4.

Summarizing, in Bucks granodiorite and in the Enchanted Rock batholith, like petrographic types are of similar thorium and uranium contents. In the Idaho batholith and probably in the Cathedral Peak and Pikes Peak granites, apparently very similar petrographic types vary widely in uranium content and vary slightly in thorium content. In the Enchanted Rock batholith, the Marysville stock, and, to a lesser extent, in the Pikes Peak granite, variations in thorium and uranium are accompanied by petrographic or textural variations;

(this does not imply, however, a necessary genetic relationship between uranium or thorium and the petrographic variation).

Where large variation in uranium occurs without concomitant gross petrographic change or thorium variation, it is highly possible that uranium may be out of secular radioactive equilibrium. Only sample F 10-24, from among the samples discussed in the immediately preceeding sections, has been checked by direct uranium chemical analysis and clearly shown to be in equilibrium.

It must be tentatively concluded, in accordance with my conclusion from analysis of the literature, that there are variations of thorium and uranium within plutons which cannot be entirely attributed to, or even related to, gross petrographic variations. Thus, in attempting to compare different types of plutons and general lithologic types, a certain element of variation, due to position within the pluton, must be expected.

RESULTS

ANALYSIS OF DATA

Radiometric data will be compared first within themselves and then with petrographic data, primarily by means of a series of scatter diagrams. Average petrographic and radiometric data of groups of samples, classified by various criteria, will then be presented (see table 16).

Relations between uranium, thorium, and Th/U ratio are shown in figures 11-18. Figures 11, 12, and 13 include samples analyzed in the course of this investigation; these are summarized in table 14. Uranium and thorium values from the literature (see tables 6 and 8) are plotted against Th/U ratio in figures 14 and 15. Figures 16, 17, and 18 show the two groups of data combined.

These two entirely independent groups of data (from the present study and from the literature) yield scatter diagrams with very similar point distributions. Thorium and uranium, plotted against one another, form trends which indicate more rapid increase of thorium than of uranium with increasing amounts of uranium or thorium in granitic rocks. Note the scatter of high-thorium points to the right of the main linear trend in figure 11. Figure 16 is a logarithmic plot, and the position of the straight trend line indicates increasing thorium relative to

uranium. Thorium, from both groups of data, forms more linear trends, when plotted against Th/U ratio, than does uranium.

Scatter diagrams of potassium versus uranium, thorium, and Th/U ratio are presented in figures 19, 20, and 21, respectively. These diagrams are very similar to the plots of thorium versus uranium. Uranium appears to increase slightly with increasing potassium content, but the points are very diffuse. Thorium increases at an increasing rate relative to potassium; the trend is fairly strong. With increasing potassium the Th/U ratio increases slightly.

If potassium is considered to be an index of evolution or of general progressive differentiation within the granitic rocks, then thorium seems to increase at a more rapid rate and more regularly with differentiation than does uranium.

Per cent standard deviations for the gamma-spectrometer data in table 14 have been calculated in order to isolate levels of variability. The standard deviations in per cent of average value are as follows: thorium, 76 per cent; uranium, 35.3 per cent; potassium, 37.4 per cent; and Th/U ratio, 83.5 per cent. Note that the standard deviation of thorium is much higher than that of uranium. It is possible that thorium values over 20

ppm are caused by secondary enrichment in thorium. If these values are excluded, the standard deviation calculated for the remaining 41 samples is lowered considerably (to 43.6 per cent) but is still higher than that of uranium. It is rather remarkable that the standard deviation of uranium, a trace element, is about the same as that of potassium, a major constituent of the rocks.

Potash-feldspar/plagioclase ratio (denoted as "R" henceforth in this paper), quartz content, per cent anorthite in plagioclase, and color index (as defined by Shand, 1947: total percentage of all minerals except plagioclase, potash feldspar, and quartz) have been derived from modal analyses (see table 15). These "petrographic parameters" are plotted against uranium, thorium, and Th/U ratio in figures 22-30. The petrographic parameters are considered to be indices of general petrogenetic evolution of granitic rocks in somewhat the same manner as potassium. The ratio R is the major criterion for petrographic classification of the granitic rocks. The relations of the average values of both petrographic parameters and radiometric data to R are summarized in figure 31. Relations which are obscure in the scatter diagrams because of diffuseness of points are more obvious when plotted as the averages of many measurements.

Figure 22 is a logarithmic plot of thorium and

uranium against R. The trend lines are straight, but they indicate variations which are not linear. The figures along the trend lines are slopes calculated graphically. According to figure 22, in rocks with progressively higher R ratios, R increases at an increasing rate relative to uranium content but stays proportional to content of thorium. Thus, thorium increases more rapidly with increasing alkalinity (R) of these granitic rocks than does uranium. Figure 22 is similar to the plots of uranium and thorium versus potassium (figures 19 and 20).

A slight increase in Th/U ratio with increase in R is shown in figure 23; however, the scatter of points is very diffuse. The trend is shown much more clearly in figure 31. Figure 31 also shows thorium content clearly increasing more rapidly than uranium with increasing R.

Relationships between uranium, thorium, and Th/U ratio and color index are shown in figures 24, 25, and 26. Uranium content is apparently unrelated to color index; thorium, however, quite markedly increases at an increasing rate with decreasing color index. This trend also appears as increasing Th/U ratio with decreasing color index in figure 26.

Figures 27-30 show the relations of quartz and anorthite content of plagioclase to uranium and thorium.

The uranium scatter diagrams are more diffuse than those of thorium. In figure 28, several thorium-rich granites may be discerned far to the right of the main linear trend. This spread of points is similar to the distribution in figure 10. If the thorium values above 20 ppm are ignored, the linear relationship of the remaining points is considerably strengthened. Thorium increases at increasing rate with increase in anorthite content of plagioclase. Th/U ratio yields a completely diffuse scatter when plotted against either quartz or anorthite content of plagioclase, and therefore, these scatter diagrams are not included.

Biotite content of the studied granites was plotted against thorium, uranium, and Th/U ratio; all relations were completely diffuse. The lack of relationship is to be expected from figure 31, which shows a distinct reversal in the trend of biotite content with increasing R.

Summarizing, the relationships shown on the foregoing scatter diagrams indicate that uranium and thorium generally vary systematically with potassium, R, and other petrographic parameters. Uranium trends are very diffuse but appear to be roughly linear. Thorium trends are much less diffuse and commonly appear curved. Th/U ratio plots are very diffuse, but where relations can be discerned, they show increase of Th/U ratio with increase

in uranium, thorium, potassium, R, and with decrease in color index. All of these trends are well substantiated by the averages plotted in figure 31.

The diffuseness of points in the diagrams is due partly to within-pluton variation and partly to sampling variation. Much of the scatter may be due to general imperfection of petrogenetic control of the uranium and thorium. This is not thought to be the case, however, since uranium and thorium are not significantly more variable than potassium or the various petrographic parameters tested. Granitic rocks representing different environments and types of genesis, however, may contain different amounts of thorium and uranium (major mineral components being equal). In such cases a significant amount of variation could originate at this level.

Thorium is more variable as well as more systematically variable, in most cases, than uranium. Some of the thorium variability, however, is caused by a few unusually thorium-rich granites, containing more than 20 ppm, which cause the spread of values observed in figures 10 and 28. Eight such granites have been analyzed; 6 of these are red or pink in color; 6 are porphyritic; 5 bear "abundant" allanite (and only these 5, among all the granites examined, contain "abundant" allanite), and the other three have fairly large amounts of allanite present;

and most of the 8 granites are relatively alkalic (high R). The uranium in these rocks is not abnormally high, and in one case (F 19-24) it is extremely low (0.84 ppm).

It is interesting to note, in figure 21 and generally in table 16, that the average Th/average U ratio is lower than the average Th/U ratio. This may be the result of the combination of large amounts of thorium with average or small amounts of uranium in some granites. Such granites would raise the average Th/U ratio more than the average thorium content. The fact that the discrepancy increases with increasing R, into the range of very thorium-rich granites, might support this explanation. It seems, at least in the thorium-rich granites, that the Th/U ratio is controlled more by thorium than by uranium.

Figure 32 presents Senftle and Keevil's (1947) data (see table 8). Th/U ratio decreases with increasing thorium between intermediate and granitic rocks in the majority of geographic groups. Likewise, with increase in uranium there is a decrease in Th/U ratio. This observation implies that uranium increases more than thorium between intermediate and granitic rocks in most regions. This observation also is contrary to the results obtained from the analysis of literature data (see figures 4 and 5) and from the data obtained in the

present investigation. Within the granitic rocks in figure 32, however, there is a strong linear increase of Th/U ratio with increasing thorium. Thus, within the granitic rocks the Th/U ratio increases with increase of uranium and/or thorium. This increase, found by Senftle and Keevil, is in agreement with the data presented in this paper.

Table 16 is a summary of averages of radiometric and petrographic data of samples grouped into several classes. The classification schemes represented in this table are quite varied. An attempt has been made to represent some of the criteria used in collection, mentioned earlier in this paper: geographic location (Rocky Mountains, Canadian Shield, and the West Coast); age (pre-, and post-Cambrian); genesis (rheomorphic, differentiated, and metasomatized or granitized); age relation to orogeny (post-tectonic); and major geological relations (disconformable and disharmonious). In many cases a sufficiently large number of samples meeting the requirements of the collection criteria was not available to justify calculation of averages. Some new groupings, subsequently found to have apparent significance, are included; textural properties (porphyritic and gneissic) and color (red).

Table 16 was constructed in order to find whether rocks belonging to these various classifications have significantly high or low uranium or thorium contents or Th/U ratios in addition to the effect that might be expected from mineralogical variation. Anomalous uranium and thorium contents and Th/U ratios of some groups, which are apparently out of accord with the associated gross mineralogical averages, may be discerned. For example, the pre-Cambrian group of granitic rocks has low Th/U ratio relative to its average R; the post-Cambrian group, however, appears to have a normal Th/U ratio relative to its average composition. This fact indicates that, relative to composition, the post-Cambrian rocks examined are enriched in thorium over uranium compared with the pre-Cambrian rocks. If compositional differences (mostly alkalinity) be ignored, or if the differences in mineralogy noted for these samples be considered true for all pre-, and post-Cambrian granitic rocks, then there is an absolute and a relative (to uranium) decrease in thorium with increase in time. This has been suggested by Marshall (1957).

Metasomatic or granitized rocks appear to have a low Th/U ratio relative to average R. This seems to be due to (relatively and absolutely) high uranium content since their thorium content is average. It is possible that

Table 16.- Comparison of radiometric and petrographic averages for various groupings of samples.

CLASSIFICATION	NO. OF SAMPLES	GAMMA DATA				PETROGRAPHIC DATA				
		Th/U	Th	U	Ave Th Ave U	K	R	Q	An	C.I. B1
Red	13	6.5	18	3	6	4	1.9	31	20	6 3.8
Rocky Mountains	13	6.2	14	2.7	4.7	3	0.7	27	28	9.5 5.8
Canadian Shield	7	5.5	11	1.7	6.5	2.4	0.8	22	20	6 3.6
Porphyritic or Seriate	13	5.4	16	3.2	5	3.1	0.7	30	26	7.9 4.9
Rheomorphic*	22	5.4	12	2.5	4	2.8	0.7	27	28	9.9 7.4
Pre-Cambrian	22	5.2	14	3	4.6	3.3	1.4	26	25	8.3 4.8
Gneissic	9	4.3	11	3	3.7	2.3	0.8	32	20	8.8 4
Metasomatic and/or Granitized	9	4.2	11	3.3	3.3	3	1.1	35	18	7.8
Late or Post-tectonic	20	4	12	3	4	2.8	0.4	31	26	10 5.7
Post-Cambrian	26	3.8	8.6	2.6	3.3	1.1	0.3	31	30	11 6.3
Differentiated	17	3.3	10	3.3	3	2.6	0.4	32	32	11 4.3
Disconformable and/or Discordant	10	3.0	9	2.9	3.1		0.2	34	31	11 7.9
West Coast	13	2.9	8.3	2.8	3.02	2	0.2	34	31	9 6.9
Disharmonious	10	2.8	8.5	2.2	3.9	2.1	0.3	29	33	13 4.9
AVERAGE	48	4.4	11	2.8	3.9	2.7	0.7	30	27	9.9 5.7

* This term used for all rocks not attributed to granitization or differentiation.

high uranium content indicates greater mobility and/or rates of diffusion of uranium than thorium.

The rocks of the Canadian Shield are anomalously high in Th/U ratio relative to R. This appears to be due to low uranium content, since thorium is present in average abundance. The low uranium content in central Canada may be accounted for by differential diffusion of uranium over thorium into upper layers of the crust during the extensive metamorphism which has affected the area. These upper layers have been subsequently eroded. It is also entirely possible that the region is a low uranium province for some entirely different reason. One such possible reason is that a series of exogenic cycles may have occurred in the Canadian Shield, thus providing ample opportunity for the oxidation and loss of uranium before each succeeding period of magma generation.

The petrographic differences among the other groups in table 16 are sufficiently large and varied so that it is impossible to attribute confidently differences in uranium or thorium content and Th/U ratio to group characteristics rather than to simple compositional differences. Therefore, the table is presented without further comment.

DISCUSSION

Any explanation proposed for the behavior of uranium and thorium in granitic rocks must explain three major phenomena: 1) the increase of both uranium and thorium in the normal or generalized petrogenetic evolution of the granitic rocks; 2) the apparent increase of thorium relative to uranium during this evolution; and 3) the greater and more systematic variation of thorium compared with the restricted (less than 6 ppm) and irregular distribution of uranium.

The general increase of uranium and thorium may be adequately explained by their large size and high ionic charge, which effectively preclude their entry into the lattices of the common rock-forming minerals. The two elements are so similar, when in the tetravalent state, that the variation of Th/U ratios observed here probably cannot be explained by the slight differences in chemical character which do exist.

Uranium can be oxidized to the hexavalent state, but thorium cannot exist in this valence state. Hexavalent uranium is considerably more soluble than tetravalent thorium and, furthermore, forms many very soluble compounds. If uranium can be oxidized during magmatic activity, then it will tend to enter volatile phases and escape from the magma. Uranium may tend to

follow thorium during differentiation but may be prevented from doing so by successive loss from the magma. This hypothesis is schematically presented in figure 33.

The postulated loss of uranium from the magma will explain the irregular distribution of uranium and its generally wider deviation than thorium from petrogenetic control. Slight changes in the ambient magmatic oxidation conditions may profoundly affect the petrochemistry of uranium but not significantly affect the petrographic parameters that are being used as criteria of petrogenetic control. The factors that control the loss of fluids from a crystallizing magma are very complicated and variable; thus, magmas which crystallized in different environments may have lost different amounts of uranium to the surrounding rocks. Figure 32 shows that in the late stages of differentiation there is an absolute decrease in uranium content in the rocks studied, which is most simply explained by loss of uranium from the magma.

The red color, predominant in thorium-rich, relatively uranium-poor granites, may indicate oxidizing conditions during magmatic activity and consequent loss of uranium. This is a particularly appealing explanation for a rock such as F 19-24, which has an extremely high Th/U ratio,

low uranium content, and marginal position in its batholith. According to McKelvey et al. (1955) the oxidation potential of tetravalent uranium is roughly the same as that of ferrous iron, and consequently red, ferruginous rocks may have been sufficiently oxidized that original tetravalent uranium was converted to the uranyl ion.

A loss of uranium in the later stages of magmatic activity might also explain the apparent reversal, which Senftle and Keevil's data indicate, in the change of Th/U ratio with increasing uranium and thorium.

Oxidizing conditions, relative to uranium, may not be attained in the earlier stages of differentiation nor in intermediate magmas; but, with increasing differentiation and in more acidic magmas, conditions may commonly become sufficiently oxidizing to cause uranium to be removed from the magma.

There is a possibility that the observed variation in Th/U ratio is not due to loss of uranium but due to addition of hydrothermal or deuteritic thorium, primarily in the form of allanite. This has been suggested by Hutton (1951) and by Coulomb et al. (1957). Tilton et al. (1954) have observed that the Essonville granite was an open system in respect to thorium and its decay products. If thorium is secondary, it should be variable

in abundance and should have been irregularly controlled by major petrogenesis. This might be the case in some of the very thorium-rich granites (see figures 10 and 32); but in general, thorium bears much more systematic relations to major petrographic parameters than does uranium. The source of late thorium would be as great a problem as the observed variation in Th/U ratios. Also, the fact that uranium shows an absolute decrease in the later stages of differentiation (see figure 31) cannot be accounted for by increase in thorium.

CONCLUSIONS

Several general conclusions may be deduced from this investigation:

- 1) Uranium and thorium are both controlled by the major petrogenesis of the granitic rocks to an extent heretofore not realized.
- 2) Uranium and thorium both increase with increasing alkalinity and acidity and, therefore, with general petrogenetic evolution of the granitic rocks.
- 3) Thorium generally increases more regularly and more rapidly than uranium, thus resulting in an increase of Th/U ratio, with general petrogenetic evolution of the granitic rocks.

- 4) Anomalously thorium-rich granites have been noted; their petrologic characteristics are very similar. . Most of them are red in color, porphyritic, alkalic, and allanite-bearing.
- 5) The increase of Th/U ratio may be explained by oxidation or complexing and consequent loss of uranium from the magma in the late stages of differentiation.
- 6) Plutons of different types may have characteristically high or low contents of uranium and thorium; however, gross compositional differences between classification types precludes accurate analysis of this problem at this time.

Figure 11 - Uranium content versus thorium content of granitic rocks in table 14.

Figure 12.- Thorium content vs. Th/U ratio of granitic rocks analysed in this investigation. (See table 14).

Figure 13.- Uranium content vs. Th/U ratio of granitic rocks analysed in this investigation. (See table 14).

Figure 14.— Uranium content vs. Th/U ratio of granitic rocks reported in the literature. (See table 6 and 8).

Figure 15.- Thorium content vs. Th/U ratio of granitic rocks reported in the literature. Closed circles refer to table 6; open circles refer to table 8.

Figure 16 - Uranium content vs. thorium content of granitic rocks. Open circles represent data from this study (see table 14). Closed circles represent data from the literature (see table 6)

Figure 17.- Uranium content vs. Th/U ratio of granitic rocks. Closed circles refer to literature data (table 6); crosses refer to data presented in this paper (table 14).

Figure 18.- Thorium content vs. Th/U ratio of granitic rocks. Closed circles refer to literature data in table 6, open circles refer to table 8, and crosses refer to data presented in this paper (table 14). Peripheral dotted circle indicates a metamorphic rock.

Figure 19 - Uranium content vs. potassium content of the granitic rocks analysed in this investigation (table 14)

Figure 20 - Thorium content vs. potassium content of the granitic rocks analysed in this investigation (table 14)

Figure 21.- Potassium content vs. Th/U ratio of granitic rocks analysed in this investigation.

Figure 22.- Uranium and thorium contents vs. potash feldspar/plagioclase ratio (R) of granitic rocks studied.

Figure 23.- Th/U ratio vs. R of granitic rocks studied. (See table 15).

Figure 24.- Uranium content vs. color index of granitic rocks studied. (See table 15).

Figure 25.- Thorium content vs. color index of granitic rocks studied. (See table 15).

Figure 26.- Th/U ratio vs. color index of granitic rocks studied. (see table 15).

Figure 28.- Thorium content vs. quartz content of granitic rocks studied. (See table 15).

Figure 27.- Uranium content vs. quartz content of granitic rocks studied. (See table 15).

Figure 29.— Thorium content vs. anorthite content of plagioclase in the granitic rocks studied. (See table 15).

Figure 30.— Uranium content vs. anorthite content of plagioclase in the granitic rocks studied. (See table 15).

Figure 31.- Radiometric and petrographic parameters plotted against R. These parameters have been determined from groups composed of samples divided, as indicated, according to R. The number of samples in each group is approximately equal.

Figure 32.- Uranium and thorium contents of a group of intermediate and granitic rocks from several geographic regions plotted against Th/U ratio. (From Senftle and Keevil, 1947).

Figure 33.- Schematic representation of postulated loss of uranium relative to thorium during normal differentiation.

APPENDIX

Samples studied in this investigation are arranged here in order of sequential collection number; that is, the number following the hyphen in the collection number. Some odd samples with slightly different designations are included at the end of the appendix. Modal analyses of these rocks are included in table 15.

Sp 2-4 Spavinaw granite: The relations of this body of rock are not well known; it is thought to be pre-Cambrian. The sample was collected 1/2 mile southwest of Spavinaw Dam along Route 22 in eastern Oklahoma; it is red, medium-grained granophyre or micrographic granite.

F 2-7 The Idaho Springs formation is a metasedimentary rock intruded by the Front Range pre-Cambrian granites. This sample was collected from Geneva Ridge in Section 8, T. 6 S., R. 75 W., Georgetown, Colorado. The hand specimen, taken from a massive quartzo-feldspathic segregation in strongly banded schists, is buff, fine- to medium-grained, allotriomorphic-granular to poikiloblastic textured granite. Large grains of muscovite are present.

F 10-15 The Pikes Peak granite is second from the acidic end of the Front Range pre-Cambrian complex

(Lovering and Goddard, 1951). It is discordant, massive, and uniform in composition and generally has sharp contacts. According to Boos (1954) it attained its present position by stoping. The hand specimen was taken 1/2 mile from the contact of granite with schist and 1/2 mile southeast of Shatters Crossing along Route 22. The specimen is pink, porphyritic quartz monzonite with phenocrysts of microcline up to 1 cm in length in a medium-grained, hypidiomorphic groundmass which has incipient mortar texture.

F 16-21 The Boulder Creek granite (Lovering and Goddard, 1951), or Mount Evans quartz monzonite of Boos (1941), forms stocks and batholiths which are satellitic to, and intruded by, the Pikes Peak granite. The Boulder Creek granite is generally harmonious, concordant, and gneissic. The specimen was taken from the summit of Mt. Evans and is gray, fine- to medium-grained quartz monzonite. The thin section shows considerable mortar texture and fracturing of all components except microcline.

F 19-24 The Pikes Peak granite is second from the acidic end of the Front Range pre-Cambrian complex (Lovering and Goddard, 1951) and is discordant, massive, and uniform in composition and generally has sharp contacts. According to Boos (1954) it attained its present position by

stoping. The hand specimen was taken from the contact of the Pikes Peak granite with the Boulder Creek granite on Epaulet Mountain in Section 36, T. 5 S., R. 75 W., Georgetown, Colorado. The specimen is pink, coarse-grained, porphyritic, quartz monzonite. Microcline is semi-poikilitic.

F 32-37 The Silver Plume granite (Mason Creek Pluton) Boos, 1954) is the youngest member of the Front Range pre-Cambrian complex. Plutons are concordant, harmonious, and disconformable stocks and sheets located between the Pikes Peak granite and the Idaho Springs formation. The granite is presumably late- or post-kinematic. The hand specimen was collected along U.S. Route 285, 11 miles southwest of the intersection of Route 285 with the Parmalee Road in Colorado. It is light-reddish-gray, hypidiomorphic-porphyritic, quartz monzonite with phenocrysts of microcline up to 1 cm in diameter.

CB 3-45 The Crested Butte porphyry was taken from a cliff on the southwest side of Crested Butte in Southwest Colorado. The intrusive is a member of the most acidic of a group of intrusives in the West Elk Mountains which may be related to the Laramide porphyries of the Front Range. This intrusive is either a stock or a laccolith and is disharmonious, disconformable, and

intrudes undeformed sediments. The hand specimen is from a cliff on the southwest side of Crested Butte and is a white seriate porphyry, with phenocrysts of sanidine, rounded (resorbed) quartz crystals, and some biotite and hornblende in a finely crystalline, felsophyric, mesostasis.

- Sc 1-49 The Rubideaux granite is an acidic member of the Lower Cretaceous (?) Southern California batholith. (Larsen, 1948). All members of this series are disharmonious and discordant against other members of the series but locally concordant against the adjacent metamorphic wall rocks. Larsen feels that the series is a product of differentiation. The hand specimen was taken from Victoria Hill in the neighborhood of Riverside, California; it is a greenish-gray, medium-grained, allotriomorphic granular, seriate, quartz monzonite.
- Sc 8-56 The Mt. Hole granodiorite is an intermediate member of the Lower Cretaceous (?) Southern California batholith (Larsen, 1948). All members of this series are disharmonious and discordant against other members of the series but locally concordant against the adjacent metamorphic wall rocks. Larsen feels that the series is a product of differentiation. The hand specimen is from the contact of Mt. Hole granodiorite with Mt. Woodson granodiorite in the vicinity of Swiss Dairy at

Arlington Place, California; the specimen is gray, fine- to medium-grained, biotite-hornblende granodiorite.

- Sc 12-60 The Woodson Mountain granodiorite is an intermediate member of the Lower Cretaceous (?) Southern California batholith (Larsen, 1948). All members of this series are disharmonious and discordant against other members of the series but locally concordant against the adjacent metamorphic wall rocks. Larsen feels that the series is a product of differentiation. The hand specimen was taken from the Arroyo del Torro, north of Elsinore, California, at the contact of the Woodson Mountain granodiorite with the Bedford Canyon formation; it is gray, medium-grained, hypidiomorphic-granular, seriate, biotite-hornblende-quartz tonalite. Inclusions with pleochroic haloes are abundant.
- Sc 15-63 The San Marcos gabbro is the oldest and most mafic member of the Lower Cretaceous (?) Southern California batholith (Larsen, 1948). It is disconformable, discordant, and disharmonious. The hand specimen was collected southwest of Elsinore along the Duluz Road and is dark-gray, fine-grained, massive, hornblende gabbro.
- Sc 25-72 The Roblar leucogranite is an acidic member of the Lower Cretaceous (?) Southern California batholith.

It is discordant, disharmonious, and has been interpreted as being the product of differentiation (Larsen, 1948). The hand specimen was taken 1 mile southwest of the Roblar Ranch sign on the Duluz-Fallbrook road; it is gray, massive, medium-grained, hypidiomorphic-granular, equigranular to seriate biotite granodiorite.

MW 3-76 The Cathedral Peak granite was collected from the Mt. Whitney area, California. This is part of a generally concordant (locally discordant) batholith of possible Upper Jurassic age which is continuous north of Mt. Whitney for more than 100 miles. The contacts are generally abrupt (Mayo, 1941) but locally become gradational (Bradley and Lyons, 1953). Cloos (1933) discerns disconformable structure within the igneous rock and interprets it to indicate magmatic intrusion; Bradley and Lyons (1933) feel that the batholith is the product of granitization. The hand specimen was taken 2 miles from the eastern contact of the batholith in the vicinity of Consultation Lake on the Mt. Whitney trail; it is light-gray, coarse-grained, porphyritic granite or quartz monzonite with idiomorphic phenocrysts or porphyroblasts of microcline (and some plagioclase) up to 2 inches in diameter.

- Y 1-96 The **Sentinel** granodiorite is an intermediate member of the Upper Jurassic (?) Yosemite igneous complex. It is generally discordant, disconformable, disharmonious, and post-tectonic (Mayo, 1933; Calkins, 1930; and Cloos, 1933). The hand specimen was collected at the foot of **S** ntinel Dome on the north side of Yosemite Valley and is gray, medium-grained, hypidiomorphic-granular, ideally granitic textured, biotite-hornblende tonalite.
- Y 7-102 The **Bridalveil** granite is a late, acidic member of the Upper Jurassic (?) Yosemite igneous complex. It is discordant, disconformable, and harmonious. The hand specimen was collected at the base of Bridalveil Falls in Yosemite National Park, California; it is gray, fine-grained, hypidiomorphic-granular, typically granitic textured, biotite granodiorite.
- Y 9-104 The **El Capitan** granite is an early member of the Upper Jurassic (?) Yosemite igneous complex. The pluton is disharmonious, disconformable, and generally abruptly discordant. The sample was collected at the base of El Capitan in Yosemite Valley, California; it is gray, massive, medium- to coarse-grained, hypidiomorphic-granular tonalite characterized by abundant, very large, aggregates of quartz.

- Y 24-119 The Cathedral Peak granite was collected from Yosemite Park (see Mw 3-73 for complete description of the relations of this granite). The hand specimen was collected from a blasted outcrop 2 miles west of Tuolumne store on the Tioga Road. The specimen is light gray, coarse-grained, porphyritic granite or quartz monzonite with idiomorphic phenocrysts or porphyroblasts of microcline up to 2 inches in diameter.
- Y 32-127 The Half Dome quartz monzonite is the earliest member and outer mantle of the eastern dome (named by Cloos, 1933) in the Upper Jurassic (?) igneous complex of Yosemite National Park. It is disconformable, discordant, and post-, or late-tectonic. The hand specimen was taken along the Tioga Road, 9.8 miles west of Tuolumne store in Yosemite National Park; it is gray, fine- to medium-grained, hypidiorhombic-granular, slightly seriate, biotite-hornblende tonalite. Large flakes of biotite and well twinned plagioclase give the rock a slight preferred orientation.
- P 11-141 The Bucks Granodiorite is an outlier of the Upper Jurassic (?) Sierra Nevada complex. The pluton is mainly concordant and harmonious with profound local discordance and granitization or assimilation. This

sample was taken from the Plumas-Merrimac area of California, on U.S. Route 24, two miles north of the Bridge across the North Fork of the Feather River. The sample is gray, massive, medium-grained, hypidio-morphic-granular, biotite-hornblende tonalite.

P 13-143 Same description as P 11-141; this sample was collected 2 miles further north along Route 24.

P 15-145 Same description as P 11-141; this sample was taken 4 miles further north along Route 24.

S 14-166 The Snoqualmie batholith is an Upper Miocene or younger, discordant, disharmonious intrusion. This sample was collected north of Lake Keechelus, Washington, along Route 10, 3 miles southwest of Snoqualmie Pass. The hand specimen is from the contact of the granodiorite with the Guye formation. The hand specimen is gray, massive, medium-grained, hypidiomorphic-granular, typically granitic textured, biotite-hornblende tonalite.

W 13-194 The Wallowa batholith is an Upper Jurassic (?) complex pluton which is concordant on a large scale but discordant locally. The contacts are generally abrupt, with some notable exceptions. It is generally disconformable. Krauskopf (1943) postulated a complex metasomatic-magmatic origin for the batholith. This sample was collected S 50° E of Hummingbird Hill and

immediately southeast of Looking Glass Lake in northeastern Oregon. The sample is gray, massive, medium-grained, hypidiomorphic-granular, biotite-hornblende tonalite. This particular locality has been presented by Krauskopf (1943) as an example of granitization at the borders of the batholith.

Id 7-202 This dark outer facies of the Idaho batholith (Anderson and Rasor, 1934) is believed to be synkinematic with the Nevadan orogeny (Anderson, 1952). According to Chapman et al. (1955) some unpublished dates indicate an age of 100 million years for this facies. The relations are very complicated, but the rock seems to be generally an autochthonous type of granite which is concordant, harmonious, conformable, and shows much evidence of endomorphic replacement. This sample was collected along Route 15, where it crosses the Payette River at Banks, Idaho; it is a gray, massive, medium-grained, hypidiomorphic-granular, biotite granodiorite.

M 3-205 The Marysville granodiorite forms a small, 3 square mile, stock which intrudes the Helena dolomite and the Empire formation, both of which are essentially undeformed except by the intrusion. The stock is concordant on a small scale but discordant on a large scale and is definitely disharmonious. Knopf (1950)

feels that it is an outlier of the Boulder batholith, exposed only 6 miles to the south. The hand specimen was taken from the center of the stock approximately 1 mile west of Marysville, Montana. The sample is a gray, fine-grained, hypidiomorphic-seriate granodiorite; it is almost a porphyry.

M 6-208 Marysville granodiorite, similar in all respects to M 3-205, but collected from the border of the stock at Drumlummon Mill in Marysville, Montana.

D 5-226 A sample from the intermediate red rock of the Duluth gabbro collected northeast of Enger Tower in Duluth, Minnesota from a transition between red rock and normal gabbro. The hand specimen is a red, fine grained, hypidiomorphic to panidiomorphic rock composed of euhedral plagioclase, some anhedral quartz, and oxidized mafic minerals.

D 9-230 Same collection locality as D 5-226; D 9-230 is intermediate between that sample and the gabbro. The hand specimen is a reddish, medium-grained, hypidiomorphic to panidiomorphic rock composed of euhedral plagioclase and pyroxene with intergranular quartz, chlorite, and iron oxide (?) stain.

D 14-235 Duluth granophyre. Contacts with the main mass of gabbro are apparently abrupt. This sample was collected at the Kenwood Avenue quarry in Duluth,

Minnesota; it is red, massive, fine-grained, panidiomorphic to hypidiomorphic granophyre. It is considerably altered to amorphous iron oxide(?).

D 17-238 Anorthositic gabbro from the Duluth lopolith.

This sample was taken from the west branch of Enger Circle on Skyline Drive in Duluth, Minnesota. The hand specimen is dark-gray to blue-black, coarse-grained, hornblende gabbro. The borders of crystals are commonly slightly fractured.

K 17-255 The Killarney granite is an irregular body of granitic rock in the Huronian sediments of central Canada. According to Quirke (1927, 1940) and Quirke and Collins (1930), the rock is a product of granitization. It is harmonious. The eastern contact of the granite with quartzite is completely transitional; the western contact is locally abrupt and locally transitional. The granite is massive to conformable. This sample was taken from the central part of the body at Le Hayes Point in Killarney, Ontario. The hand specimen is red, medium-grained, hypidiomorphic-granular, slightly seriate granite. The texture is rather strange; aggregates of very granular quartz are quite abundant.

A 4-259 The Adirondack "phacolithic" granite gneiss is a pre-Cambrian granite which intrudes Grenville

gneisses. This body is a phacolith with completely gradational borders into gneiss on all sides. It is gneissic, harmonious, generally concordant and conformable, and sillimanite- and garnet-bearing, particularly along the boundaries. According to M.S. Walton (personal communication) this is a product of metamorphic and metasomatic processes. The hand specimen was collected from Crane Pond, east of Paradox, New York, in the Adirondack Mountains. The hand specimen is red, slightly gneissic, medium-grained, allotriomorphic-granular, garnet-bearing, hornblende-quartz monzonite.

A 13-268 The Adirondack "red" granite generally forms concordant, conformable, harmonious bodies which commonly have gradational boundaries into adjacent gneiss and schist. This sample is the granitic end of a gneiss-granite transition along Route 9 just southeast of Cowne Point, New York. The specimen is pinkish-gray, massive, medium-grained, allotriomorphic-granular, biotite granite. The texture is not typically granitic; equigranular equant microcline and rounded quartz are characteristic.

NH 5-307 The Conway granite is an acidic end member of the Mississippian (?) White Mountain magma series of New Hampshire. Bodies of this granite occur in

discordant and generally disharmonious stocks and ring dikes with abrupt contacts (Billings, 1928 and 1945; Chapman and Williams, 1935). The granite has been interpreted by Chapman and Williams (1935) as a product of differentiation. The hand specimen was taken from Redstone quarry, south of North Conway, New Hampshire; it is red, massive, medium-grained, allotriomorphic- to hypidiomorphic-granular, biotite granite.

NH 29-331 The Mascoma Group is a member of the Upper Devonian (?) Oliverian magma series. Its plutons form the concordant, conformable, harmonious, gradationally bounded, gneiss-domes of New England (Chapman, 1939). The hand specimen was collected from a quarried block 1 mile south of Goss School in the Mascoma quadrangle, New Hampshire; it is gray, gneissic, fine- to medium-grained, granoblastic, quartz monzonitic gneiss. The texture is metamorphic, not granitic.

NH 43-345 The Bethlehem gneiss is a mafic member of the Upper Devonian (?) New Hampshire magma series. Bodies of this gneiss are generally concordant, conformable, harmonious and presumably synkinematic. Billings (1945) postulated a remobilized granitization origin for these bodies. This sample was taken along Route 10

approximately 1 1/2 miles north of North Grantham, New Hampshire; it is gray, strongly gneissic, medium-grained, allotriomorphic-granular or granoblastic, quartz monzonitic gneiss.

NH 47-249 The Concord granite is an acidic member of the Upper Devonian (?) New Hampshire magma series. This granite occurs in dike-like intrusions and stocks (Chapman, 1952; Billings, 1946). The sample was taken along Route 11, 3/4 mile south of Georges Mills, New Hampshire; it is gray, massive, fine-grained, allotriomorphic-granular, muscovite-quartz monzonite.

W 3-399 The Wichita Granophyre is a member of a pre-Cambrian stratiform complex in which granophyric material overlies gabbro and anorthosite (Hamilton, 1956). This hand specimen was collected at Elmer Thomas Dam in the Wichita Mountains of Oklahoma; it is red, massive, fine- to medium-grained, panidiomorphic to hypidiomorphic, semi-graphic granite.

E 3-402 The Enchanted Rock batholith is pre-Cambrian, slightly disharmonious, disconformable and discordant on a large scale (Hutchinson, 1956). The intrusion is apparently complex with peripheral zonation of petrographic types. This sample was taken from the porphyritic granite of the intermediate zone; it is red, massive, coarse-grained, hypidiomorphic-porphyritic,

slightly seriate biotite quartz monzonite.

E 4-403 This Enchanted Rock batholith sample is from the same location and description as sample E 3-402. The present sample was collected 100 feet from E 3-402.

E 5-404 Enchanted Rock batholith; the present sample was collected from the same outcrop as was E 3-402 but from an aplitic dike cutting the porphyritic material. The hand specimen is gray, fine-grained, allotriomorphic-granular, aplitic, biotite-quartz monzonite.

Ka 6-405 This sample of Cold Spring granite from the Cold Spring Granite Co. quarry in Rockville, Minnesota was donated by Dr. S.S. Goldich.

KA 72-406 The Northern Lights group is an autochthonous gneiss northwest of the Saganaga granite. Grout (1929 a, 1929 b) feels that this is the root zone of some of the younger granites in northern Minnesota. The hand specimen is from the Canadian Rangers cabin on the north side of the northwest channel of Northern Lights Lake, east of the portage into Saganaga Lake. This sample is a fine-grained granoblastic gneiss, and was donated by Dr. S.S. Goldich.

P 210G (407) The Saganaga granite is a Laurentian (?) semi-concordant, harmonious granite or granite gneiss which Grout (1933) feels has been exposed to a depth

of 25 kilometers. This sample was collected by Dr. S.S. Goldich from the lower part, exposed in Saganaga Lake. The sample is gray, medium-grained, hypidio-morphic-granular, hornblende tonalite.

KA 80-408 The Burntside Gneiss is an early Algonian (?) gneissic granite which is conformable, generally concordant, and harmonious. This sample was collected from a small island near the east end of Burntside Lake, Minnesota; it is a gray, massive to slightly gneissic, medium-grained, granitic gneiss.

V 1-409 The Vermillion granite is the largest batholith along the Canadian border in Minnesota. It is complex but generally harmonious. The sample was collected by Dr. S.S. Goldich near a road cut 200 feet east of the intersection of Echo trail and Sioux River, St. Louis County, Minnesota. The hand specimen is red, medium-grained, allotriomorphic-granular, slightly seriate, quartz monzonite.

KA 27-410 This sample of the Montevideo granite gneiss is from the S.E. 1/4, Section 20 on U.S. Route 212, Minnesota. The hand specimen is fine- to medium-grained, allotriomorphic seriate, granite gneiss.

M-4 Dark outer facies of the Idaho batholith (see sample Id 7-202 for a full description of the batholith). This specimen was taken on the north shore of Arrowrock

reservoir, 4 miles from the turnoff of the dam road from Route 20 east of Boise, Idaho. The sample is gray, medium-grained, hypidiomorphic-granular, biotite tonalite.

12M The Inner facies of the Idaho batholith is believed by Anderson (1942, 1948) to be post-orogenic to the Nevadan disturbance, but there is considerable doubt as to its absolute age. It is complex but has been considerably altered. This hand specimen was collected on the north shore of Arrowrock reservoir, 12 miles from the turnoff of the dam road from Route 20 east of Boise, Idaho; it is gray, medium-grained, allotriomorphic-granular, leucogranodiorite. There is considerable mortar texture.

JW 70 This is an aggregate of 4 samples of the Bonsal tonalite member of the Lower Cretaceous (?) Southern California batholith. Its plutons are generally discordant, disharmonious, and disconformable. The plutons are characterized by elongate inclusions and schlieren. All samples are gray, medium-grained, hypidiomorphic-seriate, biotite-hornblende tonalite.

JW 32 This is an aggregate of several samples of the Mt. Morrison gneiss member of the Front Range igneous complex. There is considerable controversy over whether this is a para-gneiss or an ortho-gneiss. It is

harmonious and autochthonous. Texture is medium grained, and gneissic.

CITED REFERENCES

- Adams, J.A.S., and Maeck, W.J., 1954, Fluorometric and colorimetric microdetermination of uranium in rocks and minerals: *Anal. Chem.*, v. 26, p. 1635-1639.
- Adams, J.A.S., J.E. Richardson, and C.C. Templeton, Determination of thorium and uranium by gamma-ray spectral analysis compared with an alpha activity-fluorometric uranium method, (preprint).
- Ahrens, L.H., 1957, A survey of the quality of some of the principle abundance data of geochemistry, in Ahrens, L.H., F. Press, K. Rankama, and S.K. Runcorn (editors), 1957, *Physics and chemistry of the earth*, v. 2, pp. 30-45.
- Anderson, A.L., 1942, Endomorphism of the Idaho batholith: *Bull. GSA*, v. 53, p. 1099-1126.
- Anderson, A.L., 1948, Role of the Idaho batholith during the Larimide orogeny: *Econ. Geol.*, v. 43, p. 84-99.
- Anderson, A.L., 1952, Multiple emplacement of the Idaho batholith: *Jour. Geol.*, v. 60, p. 255-265.
- Anderson, A.L. and Rasor, A.C., 1934, Composition of a part of the Idaho batholith in Boise County, Idaho: *Am. Jour. Sci.*, v. 27, p. 287-294.
- Asayama, T., 1936, *Jap. Jour. Astron. Geoph.*, v. 14, p. 19-26.
- Asayama, T., 1954, The radium content and chemical composition of granitic rocks in Japan, especially in the Tanakami-Mikumo and the Hiei regions, Shiga and Kyoto Prefectures: *Memoir of the Faculty of Industrial Arts, Kyoto Technical University, Science and Technology*, vol. 3, (B).
- Barth, T.J.W., 1838, Radium and the petrology of certain granites of Finland: *Am. Jour. Sci.*, v. 235a, p. 231-245.
- Billing, M.P., 1928, The petrology of the North Conway quadrangle in the White Mountains of New Hampshire: *Am. Acad. Arts Sci. Proc.*, v. 63, p. 67-137.
- Billings, M.P., 1945, Mechanics of igneous intrusion in New Hampshire: *Am. Jour. Sci.*, v. 243a, p. 40-66.

- Billings, M.P., and Keevil, N.B., 1946, Petrography and radioactivity of four Paleozoic magma series in New Hampshire: Bull. GSA, v. 57, p. 797-827.
- Boos, M.F., 1954, Genesis of pre-Cambrian granitic pegmatites in the Denver Mountain Parks area, Colorado: Bull. GSA, v. 65, p. 115-142.
- Borkowski, C.J., and R.L. Clark, 1953, Gamma-ray energy resolution with NaI-TlI scintillation spectrometers: Rev. Sci. Instr., v. 24, p. 1040-1050.
- Bradley, C. and E.J. Lyons, 1953, A mode of evolution of a granitic texture: GSA Mem. 52, p. 101-110.
- Brown, H., W.J. Blake, A.A. Chodos, R. Kowalkowski, C.R. McKinney, G.J. Neuerburg, L.T. Silver, and A. Uchiyama, 1953, Leaching studies of interstitial material in igneous rocks (abstract): Bull. GSA, v. 64, p. 1400.
- Brown, Harrison and L.T. Silver, 1956, The possibilities of obtaining long range supplies of uranium, thorium, and other substances from igneous rocks: U.S. Geol. Survey Prof. Paper 300, p. 91-95.
- Calkins, F.C., 1930, The granitic rocks of the Yosemite Region, in Matthes, F.E., Geologic history of the Yosemite Valley, U.S. Geol. Survey Prof. Paper 160, p. 120-128.
- Chapman, C.A., 1939, Geology of the Mascoma quadrangle, New Hampshire: Bull. GSA, v. 50, p. 127-180.
- Chapman, C.A., 1952, Structure and petrology of the Sunapee quadrangle, New Hampshire: Bull. GSA, v. 63, p. 381-425.
- Chapman, R.W., 1955, Distribution of uranium in the Boulder batholith, Montana: in Geological Investigations of Radioactive Deposits, T E I 590, p. 312-314.
- Chapman, R.W., R. Gottfried, C.L. Waring, 1955, Age determinations from the Boulder batholith and other batholiths of western Montana: Bull. GSA, v. 66, p. 607-610.

- Chapman, R.W. and C.R. Williams, 1935, Evolution of the White Mountain magma series: *Am. Mineralogist*, v. 20, p. 502-530.
- Cloos, E., 1933, Structure of the Sierra Nevada batholith: 16th Int. Geol. Cong., Guidebook 16, p. 40-45.
- Coppens, Rene, 1951, Etude de radioactivite de quelque par emulsion photographique, *Bull. Soc. Franc. Mineral*, vol. 73, p. 217-321.
- Coulomb, R., M. Goldstein, M. Le Mercier, L'Uranium dans quelques granites Francais: (preprint).
- Davidson, C.F., 1951, The distribution of radioactivity: *Mining Mag. (London)*, v. 85, p. 329-340.
- Davidson, C.F., 1957, Radiogeology in Russia: The *Mining Magazine*, November, p. 3-11.
- Deutsch, S. and Picciotto, F., 1950, The presence of uraninite in the accessory minerals of the Baveno granite: *Experientia*, v. XII, p. 333.
- Evans, R.D. and Clark Goodman, 1941, Radioactivity of rocks, *Bull. GSA*, v. 52, p. 459-490.
- Evans, R.D. and R.W. Raitt, 1935, The radioactivity of the earth's crust and its influence on cosmic-ray electroscope observations made near ground level: *Phys. Rev.*, v. 48, p. 171-176.
- Fano, Ugo, 1953, Gamma-ray attenuation: *Nucleonics*, v. 11, p. 8-12, 55-61.
- Faul, Henry (editor), 1954, *Nuclear Geology*, John Wiley & Sons, New York.
- Finney, G.D. and R.D. Evans, 1935, The radioactivity of solids determined by alpha-ray counting: *Phys. Rev.*, v. 48, p. 503-511.
- Friedlander, G. and J.W. Kennedy, 1949, *Nuclear and Radiochemistry*, John Wiley & Sons, New York.
- Goodman, C. and R.D. Evans, 1941, Age measurements by radioactivity: *Bull. GSA*, v. 52, p. 491-544.

- Gottfried, David and E.S. Larsen, 1954, Distribution of uranium in igneous complexes: in Geological Investigations of Radioactive Deposits, TEI-440, p. 202-205.
- Grimaldi, F.S., I. May, and M.H. Fletcher, 1952, U.S. Geological Survey fluorimetric methods of uranium analysis: U.S. Geol. Survey Circ. 199, 20 pp.
- Grimaldi, F.S., I. May, M.H. Fletcher, and Jane Titcomb, 1954, Collected Papers on methods of analysis for uranium and thorium: U.S. Geol. Survey Bull. 1006, 184 pp.
- Grout, F.F., 1929a, The Saganaga granite, Minnesota-Ontario: Jour. Geol. v. 37, p. 582-591.
- Grout, F.F., 1929 b, Ages and differentiation of batholiths near the Minnesota-Ontario boundary: Bull. GSA, v. 40, p. 791-809.
- Grout, F.F., 1933, Structural features of the Saganaga granite of Minnesota-Ontario: 16th Inter. Geol. Cong. Rept., v. 1., p. 255-270.
- Hadley, J.B., 1942, Stratigraphy, structure, and petrology of the Mt. Cube area, New Hampshire: Bull. GSA, v. 53, p. 112-176.
- Hamilton, Warren B., 1956, Precambrian rocks of Wichita and Arbuckle Mountains, Oklahoma: Bull. GSA, v. 57, p. 1319-1330.
- Hatuda*, Z., 1934, Mem. Coll. Sci., Kyoto Univ., (B), v. 10, p. 63-72.
- Hatuda, Z., 1936, Radium content of rocks of Nippon: Mem. Coll. Sci., Kyoto University, (B), v. 12, p. 1-18.
- Hayase, Ichikazu*, 1955, The radioactivity of rocks and minerals studied with nuclear emulsions: III Radioactivity of biotite of the Tanakamika granite, Shiga Prefect, Japan: Mem. Coll. Sci. Univ. Kyoto, Sec. B, v. 22, P. 177-184.
- Hee, A. Mme., R.P. Derville, and M. Jarovoy, 1954, Determination of radioactivity of the quincy granite by the photographic method: Am. Jour. Sci., v. 252, p. 736-744.

- Hofstadter, R. and J.A. McIntyre, 1950, Gamma-ray spectroscopy with crystals of NaI (Tl): *Nucleonics*, v. 7, p. 32-37.
- Hollander, J.M., Perlman, I., and Seaborg, G.T., 1953, Table of isotopes: *Rev. Modern Physics*, v. 25, p. 469-691.
- Holmes, Arthur, 1926, Contributions to the theory of magmatic cycles: *Geol. Mag.*, v. 63, p. 306-329.
- Holmes, Arthur, 1937, The origin of primary lead ores: *Econ. Geol.*, v. 32, p. 763-782.
- Holmes, Arthur, 1938, The origin of primary lead ores: II: *Econ. Geol.*, v. 33, p. 829-867.
- Hurley, P.M., 1950, Distribution of radioactivity in granites and possible relation to Helium age measurement: *Bull. GSA*, v. 61, p. 1-8.
- Hurley, P.M., 1954, The helium age method and the distribution and migration of helium in rocks: in Faul, Henry, (editor): *Nuclear Geology*, John Wiley & Sons, New York, p. 301-328.
- Hurley, P.M., 1956, Direct radiometric measurement by gamma-ray scintillation spectrometer: *Bull. GSA*, v. 67, p. 395-412.
- Hurley, P.M. and H.W. Fairbairn, 1957, Abundance and distribution of uranium and thorium in zircon, sphene, apatite, epidote, and monazite in granitic rocks: *Trans. Amer. Geophys. Union*, v. 38, p. 939-944.
- Husmann, Otto, 1956, Determination of the thorium and uranium content of gneisses and of anatectic and magmatic rocks of the central Black Forest by means of coincidence method: *Neues. Jahrb. Mineral., Monatsh.*, v.5, p. 108-120.
- Hutchinson, R.M., 1956, Structure and petrology of Enchanted Rock batholith, Llano and Gillespie Counties, Texas: *Bull. GSA*, v. 67, p. 763-806.
- Hutton, C.O., 1951, Allanite from Wilmot Pass, Fiordland, New Zealand: *Am. Jour. Sci.*, v. 249, p. 208-214.

- Ingham, W.N. and N.B. Keevil, 1951, Radioactivity of the Bourlamaque, Elzevir, and Cheddar batholiths: Bull. GSA, v. 62, p. 131-148.
- Jeffreys, H., 1936, Gerl. Beitr. Geophys., v. 47, p. 149-171.
- Joly*, J., 1924, Uber die Radioaktivitat der Gestein: Naturw., v. 12, p. 693-699.
- Kahn, B. and Lyon, W.S., 1953, Scintillation spectrometer in radiometric analysis: Nucleonics, v. 11, p. 61-63.
- Kallmann, H., M. Furst, and M. Sidran, 1952, Scintillation counting techniques: Nucleonics, v. 10, p. 15-18.
- Keevil, N.B., 1938 a, Th/U ratios of rocks and their relation to lead ore genesis: Econ. Geol., v. 33, p. 685-696.
- Keevil, N.B., 1938 b, Distribution of helium and radioactivity in rocks: Part I. Mineral separates from the Quincy granite: Am. Jour. Sci., v. 36, p. 406-416.
- Keevil, N.B., 1943, The distribution of helium and radioactivity in rocks, V: Rocks and associated minerals from Quebec, Ontario, Manitoba, New Jersey, New England, New Brunswick, Newfoundland, Tanganyika, Finland, and Russia: Am. Jour. Sci., v. 241, p. 277-306.
- Keevil, N.B., 1944, Th-U ratios in rocks and minerals: Am. Jour. Sci., v. 242, p. 309-321.
- Keevil, N.B., A.R. Keevil, W.N. Ingham, and G.P. Crombie, 1943, Causes of variation in radioactivity data: Am. Jour. Sci., v. 241, p. 345-365.
- Keevil, N.B., E.S. Larsen, and F.J. Wank, 1944, Distribution of helium and radioactivity in rocks, VI: The Ayer granite-migmatite at Chelmsford, Mass.: Am. Jour. Sci., v. 242, p. 345-353.
- Knopf, Adolph, 1950, The Marysville granodiorite stock, Montana: Am. Mineralogist, v. 35, p. 834-844.

- Krauskopf, K.B., 1943, The Wallowa batholith: Am. Jour. Sci., v. 241, p. 607-621.
- Krylov, A. Ya., 1956, Distribution of U and Th in the granite massifs of Dzheta-oguz and Alabash: Trudy Radiovogo Inst. Im. V.G. Khlopina, Khim. i Geokhim., v. 7, p. 200-213.
- Larsen, E.S., 1948, Batholith and associated rocks of Corona, Elsinore, and San Luis Rey quadrangles, Southern California: GSA Mem. 29.
- Larsen, E.S. and N.B. Keevil, 1942, The distribution of helium and radioactivity in rocks, III: Radioactivity and petrology of some California intrusives: Am. Jour. Sci., v. 240, p. 204-215.
- Larsen, E.S. and N.B. Keevil, 1947, Radioactivity of the rocks of the batholith of Southern California: Bull. GSA, v. 58, p. 483-493.
- Larsen, E.S., Jr., George Phair, David Gottfried, and W.L. Smith, 1956, Uranium in magmatic differentiation: U.S. Geol. Survey Prof. Paper 300, p. 65-74.
- Loevinger, R. and M. Berman, 1951, Efficiency criteria in radioactivity counting: Nucleonics, v. 9, p. 26-39.
- Lovering, T.S. and E.N. Goddard, 1951, Geology and ore deposits of the Front Range, Colorado: U.S. Geol. Survey Prof. Paper 223, p. 23.
- Marshall, R.R., 1957, Isotopic composition of common leads and continuous differentiation of the crust of the earth from the mantle: Geochim. et Cosmochim. Acta, v. 12, p. 225-237.
- Mayo, E.B., 1935, Some intrusions and their wall rocks in the Sierra Nevada: Jour. Geol., v. 43, p. 673-689.
- Mayo, E.B., 1941, Deformation in the interval Mt. Lyell-Mt. Whitney, California: Bull. GSA, v. 52, p. 1001-1084.
- McKelvey, V.E., D.L. Everhart, and R.M. Garrels, 1955, Origin of uranium deposits: Fiftieth Anniversary Volume of Econ. Geol., p. 464-533.

- Moteff, John, 1955, Tenth-value thicknesses for gamma-ray absorption: Nucleonics data sheet no. 5, Nucleonics.
- Neuerburg, G.J., 1956, Uranium in igneous rocks of the United States: U.S. Geol. Survey Prof. Paper 300, p. 55-64.
- Nier, A.C., 1938, The isotopic constitution of lead from various sources: J. Am. Chem. Soc., v. 60, p. 1571.
- Nier, A.C., 1950, A redetermination of the relative abundances of the isotopes of carbon, nitrogen, oxygen, argon and potassium: Phys. Rev., v. 77, p. 789-793.
- Phair, George, 1956, Distribution of uranium in igneous complexes: in Geological Investigations of Radioactive Deposits: TEI 620, p. 310-318.
- Phair, George and David Gottfried, 1955, Distribution of uranium in igneous complexes: in Geological Investigations of Radioactive Deposits: TEI 590, p. 310-312.
- Picciotto, E.E., 1950, Distribution de la radioactivite dans les roches eruptives: Soc. Belge Geologie Bul., v. 59, p. 170-198.
- Piggot, C.S., 1931, Radium in Rocks: II. Granites of eastern North America from Georgia to Greenland: Am. Jour. Sci., v. 21. p. 28-36.
- Piggot, C.S. and H.E. Merwin, 1932, Radium in rocks; IV: Am. Jour. Sci., v. 23, p. 49-56.
- Piggot, C.S., 1938, Radium in rocks: The radium content of four groups of Pre-Cambrian granites of Finland: Am. Jour. Sci., v. 235 a, p. 28-36.
- Poole*, J H.J. and J. Joly, 1924, The radioactivity of basalts and other rocks: Phil. Mag., v. 55, p. 819.
- Quirke, T.T., 1927, Killarney gneiss and migmatites: Bull. GSA, v. 38, p. 753-766.
- Quirke, T.T., 1940, Granitization near Killarney Ontario: Bull. GSA, v. 51, p. 237-254.

- Raitt*, R.W., 1935, Alpha ray counting: Ph.D. Thesis, Calif. Inst. Tech., p. 1-78.
- Rankama, Kalervo, 1954, Isotope Geology, McGraw-Hill, New York.
- Rankama, Kalervo and Sahama, Th.G., 1950, Geochemistry: Univ. of Chicago Press, Chicago.
- Senftle, F.E. and N.B. Keevil, 1947, Thorium-uranium ratios in the theory of genesis of lead ores: Trans. Am. Geophys. Union, v. 28, p. 732-738.
- Shand, S.J., 1947, Eruptive rocks, John Wiley & Sons, New York.
- Slack, H.A. and K. Whitham, 1951, A further investigation of the radioactivity of the Round Lake and Elzevir batholiths: Trans. Am. Geophys. Union, v. 32, p. 44-48.
- Smith, W.L. and F.J. Flanagan, 1956, Use of statistical methods to detect radioactivity change due to weathering of a granite: Am. Jour. Sci., v. 254, p. 316-324.
- Smith, W.L., M.L. Frank, and A.M. Sherwood, 1957, Uranium and thorium in the accessory allanite of igneous rocks: Am. Min., v. 42, p. 367-378.
- Soloviev, S.P., 1936, The granodiorite laccolith of the Malka River (North Caucasus) and its content of radium: Am. Jour. Sci., v. 32, p. 380-391.
- Szadeczky-Kardoss*, E., 1953, The geochemical migration of the elements: II, The segregation of magmatic provinces: Acta. Geol. Acad. Sci. Hung., v. 2, p. 145-167.
- Tauson, L.V., 1956, The geochemistry of uranium in granitoids of the Chernovinsk Massif (Atlas Mts.): Geokhimiya, no. 3, p. 9-17.
- Tauson, L.V., B.I. Zlobin, and L.L. Lenova, 1956, Distribution of uranium in granitoid complex of the Sysmyr batholith (Central Tien Shan): Geokhimiya, no. 7, p. 11-19.

- Tilton, G.R., C.C. Patterson, H. Brown, M. Inghram, R. Hayden, D. Hess, and E.S. Larsen, Jr., 1954, The isotopic composition and distribution of lead, uranium and thorium in a pre-Cambrian granite: U.S. Atomic Energy Commission, AECU 2840.
- Trudy Radievogo Instituta im. V.G. Khlopina.* Moscow (Academy of Sciences) v. 5 (2), 292 pp.
- Trudy Radievogo Instituta im. V.G. Khlopina.* Moscow (Academy of Sciences) v.7, 216 pp.
- Urry*, W.D., 1936, Determination of the thorium and radium contents of rocks; J. Chem. Phys., v. 4, p. 34-48.
- Walton, Matt., 1955, The emplacement of "granite": Am. Jour. Sci., v. 253, p. 1-18.
- Wendt, I, 1955, Bestimmung von Thorium, Uran, und Kalium an unaufgeschlossenen Gesteinen mit dem Zählrohr: Dissertation, Gottingen.

* references which were not consulted.