Physics

This document consists of 30 pages.
Series A.

UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY

DIRECT RADIOMETRIC MEASUREMENT OF THE URANIUM AND THORIUM SERIES IN EQUILIBRIUM BY GAMMA-RAY SCINTILLATION SPECTROMETER*

By

Patrick M. Hurley

February 1955

Trace Elements Investigations Report 499

This preliminary report is distributed without editorial and technical review for conformity with official standards and nomenclature. It is not for public inspection or quotation.

---

*This report concerns work done on behalf of the Division of Research of the U. S. Atomic Energy Commission.

USGS - TEI-499

PHYSICS

Distribution (Series A)	No. of copies
Argonne National Laboratory	1
Atomic Energy Commission, Washington	2
Battelle Memorial Institute, Columbus	1
Carbide and Carbon Chemicals Company, Y-12 Area	1
Division of Raw Materials, Albuquerque	1
Division of Raw Materials, Butte	1
Division of Raw Materials, Casper	1
Division of Raw Materials, Denver	1
Division of Raw Materials, Hot Springs	1
Division of Raw Materials, Ishpeming	1
Division of Raw Materials, Phoenix	1
Division of Raw Materials, Richfield	1
Division of Raw Materials, Salt Lake City	1
Division of Raw Materials, Washington	3
Division of Research, Washington	1
Dow Chemical Company, Pittsburg	1
Exploration Division, Grand Junction Operations Office	1
Grand Junction Operations Office	1
National Lead Company, Winchester	1
Technical Information Service, Oak Ridge	6
Tennessee Valley Authority, Wilson Dam	1
U. S. Geological Survey:	
Fuels Branch, Washington	1
Geochemistry and Petrology Branch, Washington	15
Geophysics Branch, Washington	1
Mineral Deposits Branch, Washington	1
E. H. Bailey, Menlo Park	1
A. L. Brokaw, Grand Junction	1
N. M. Denson, Denver	1
C. E. Dutton, Madison	1
W. L. Emerick, Plant City	1
A. H. Koschmann, Denver	1
R. A. Laurence, Knoxville	1
J. D. Love, Laramie	1
L. R. Page, Washington	1
P. C. Patton, Denver	1
J. F. Powers, Salt Lake City	1
C. B. Read, Albuquerque	1
Q. D. Singewald, Beltsville	1
A. E. Weissenborn, Spokane	1
TEPCO, Denver	2
TEPCO, RPS, Washington, (including master)	2
	65

CONTENTS

Page

Abstract 4
Introduction 4
Instrumentation 10
Relative spectra of the uranium and thorium series 13
Method of analysis and tests 17
Source geometry 24
Acknowledgments 29
References 30

ILLUSTRATIONS

Figure 1. Detail of photomultiplier, crystal source holder, and shield 11

2. Two-channel scintillation spectrometer 12

3. Gamma scintillation spectrum for uranium series in equilibrium 14

4. Gamma scintillation spectrum for thorium series in equilibrium 15

5. Comparison of U and Th series spectra below 0.4 mev showing bands used for Th/U analysis 16

6. Factors in selection of optimum channel width for thorium discrimination 18

7. Uranium, thorium, and U/Th as functions of R_V, Q_T .. 22

8. Estimated precision for different proportions of uranium and thorium, for single comparison against standard 25

9. Deviation from normal value of count per unit sample weight, as function of total sample weight, showing absorber end effect 27

TABLES

Table 1. Principal gamma rays in the uranium and thorium series 8

2. Results of the method on a number of test samples ... 26

DIRECT RADIOMETRIC MEASUREMENT OF THE URANIUM AND THORIUM SERIES IN EQUILIBRIUM BY GAMMA-RAY SCINTILLATION SPECTROMETER

By Patrick M. Hurley

ABSTRACT

When uranium and thorium are in secular equilibrium with their decay products, these elements may be determined by this direct radiometric method if the materials show more than 0.01 percent equivalent uranium. This method is based on the measurement in a scintillation spectrometer of the relative amplitude of the response from the 238 kev gamma ray from Pb^{212}. Tests on known samples containing different relative proportions of uranium and thorium have verified the predicted precision of better than 4 percent standard deviation per single comparison with a standard when the element measured is a major contributor to the activity.

INTRODUCTION

The scintillation spectrometer may be used for radiochemical analysis (Kahn and Lyon, 1953) if the material to be analyzed is not too complicated by unknown proportions of different gamma-ray emitters. When there are different gamma-ray emitters present, the analysis generally requires a separation of photoelectric absorption and counting efficiency from other absorption processes, and the calibration in absolute terms becomes difficult. When the uranium or thorium series is in equilibrium this difficulty is eliminated because the proportion of gamma emitters remains constant, and an analysis for either uranium or thorium may be made on the basis of total counts in any part of the spectrum as long as source geometry and absorption is held constant or corrections for variations are made. This makes possible the discrimination of the thorium and uranium series by counting two bands in the spectrum that are selected to give a ratio that varies for different proportions of the two series in the sample.

There is a considerable need for a rapid radiometric measurement of uranium in the presence of thorium and vice versa in mineral samples, and although the requirement that the two series must be in secular equilibrium reduces the general usefulness to some extent, by far the major proportion of radioactive mineral occurrences are in equilibrium or so close to it that no important error results from assuming that equilibrium exists. In the uranium series 95 percent of equilibrium is established in 3.5×10^5 years and in the thorium series only 30 years are required. Emanation leakage may keep the mineral out of equilibrium and cause an error not only in the abundance of the parent but also in the ratio of the uranium to thorium because thoron will not diffuse as far as radon owing to its shorter life. Generally this error is not significant, and an estimate of its possible importance can be made from experience depending on the physical nature and history of the mineral aggregate. Chemical leaching of the daughter elements is more difficult to estimate.

In this investigation the materials were limited to those in which the content of the uranium and its products, or its thorium equivalent in activity, exceeded 0.01 percent. Sufficient work has been done to indicate that the same technique can be extended to geological materials containing as little as a few parts per million equivalent uranium with the use of a modified source. This latter technique is applicable to common rocks but requires source absorption correction and discrimination of potassium. It will be described separately in a subsequent report.

The method that is here described has several advantages if the precision attainable is acceptable to the user. Source absorption can be made negligible for materials of ore grade which is one of the biggest problems in methods using the discrimination of alpha particles (Pierson, 1951). The method proposed by Eichholz, Hilborn and McMahon (1953) using combined beta-gamma count eliminates the problem of equilibrium and looks very good. Information on the precision of the method for mixtures of uranium and thorium is not given in sufficient detail to make a comparison with the method proposed here. Larger samples can be used than are possible in the beta-gamma method for materials low in these elements. This has an important advantage in the assay of granites, for example, in which much of the activity occurs in small accessory minerals and there is great difficulty in obtaining a small representative sample of the whole rock. Another advantage is that the method is quite rapid for routine work. As precision is gained by replicate comparisons with a single standard, the assay time will depend on the precision specifications.

The development of scintillation spectrometry has been described adquately in the physical literature (Hofstadter and McIntyre, 1950). It may be helpful to include a brief statement of the principles of the gamma energy discrimination process at this time, as the subject has not been introduced to any extent in the geological literature. The principle of scintillation counters is well known in geology as they are in common use in the exploration for uranium. Gamma rays acting on the scintillation crystal may give up all or some of their energy in the crystal. When the gamma ray gives up its entire energy, a pulse of light proportional to the energy of the gamma may result. This occurs for the most part when photoelectrons are produced which do not emerge from the crystal. An imperfect line spectrum of light pulses is then superimposed on a continuous spectrum of pulses resulting from gamma rays which have lost only part of their energy by Compton scattering or resulting from photoelectrons that have emerged from the crystal. Discrete spectra may also result from higher energy gamma rays producing electron pairs. These pulses are reduced in magnitude by an amount equivalent to at least 1.02 mev which is the energy required to form the electron pair.

The abundance of gamma rays of different energy resulting from the breakdown of the uranium and thorium series is indicated by the partial list in table 1. Only the prominent ones emerge as peaks above the base spectrum.

The light pulses in the crystal are transformed linearly into voltage pulses by a photomultiplier tube and amplifier. These pulses are fed into an analyzer in which an adjustable base level threshold eliminates all pulses below a certain amplitude. Riding above this base line threshold is another adjustable voltage level above which all pulses are eliminated. The instrument therefore provides a channel, or gate, of adjustable width which can be set at any voltage and which can be used to scan the spectrum of pulses. The pulses passing through the gate are recorded on a scaler or rate meter. More than one analyzer channel may be fed the same signal of pulses, thereby counting different parts of the

Table 1.--Principal gamma rays in the uranium and thorium series.1/

Z	Isotope	A	Energy in mev of gamma rays
			Uranium 238 series
92	Uranium	238	~0.05 Coincident with 22 percent of alpha particles
90	Thorium	234	0.093 20 percent of disintegrations
91	Protactinium	234	0.82 Weak
92	Uranium	234	$\gamma_1 0.053$, $\gamma_2 0.093$, $\gamma_3 0.118$ $\gamma_1/\gamma_2/\gamma_3 = 1/\sim 0.2/0.4$
90	Thorium	230	0.068, (0.14, 0.24) e/γ ~46 with 25 percent of alpha particles
88	Radium	226	0.188 e/γ = 0.5 to 0.9 with 5.7 percent of alpha particles
86	Emanation	222	
84	Polonium	218	
82	Lead	214	$\gamma_1 0.053$, $\gamma_2 0.242$, $\gamma_3 0.257$, $\gamma_4 0.295$, $\gamma_5 0.352$ $\gamma_2/\gamma_4/\gamma_5 = 0.2/0.55/1.0$
83	Bismuth	214	$\gamma_1 0.609$, $\gamma_2 0.766$, $\gamma_3 0.933$, $\gamma_4 1.120$, $\gamma_5 1.238$, $\gamma_6 1.379$, $\gamma_7 1.520$, $\gamma_8 1.761$, $\gamma_9 1.820$, $\gamma_{10} 2.200$, $\gamma_{11} 2.420$ in ratio 9/1.3/1.1/2.6/1.0/0.9/0.7/3.2/0.2/1.0/0.5 with β^-
84	Polonium	214	
82	Lead	210	0.0467 (3 percent) (e/γ ~ 20)
83	Bismuth	210	No γ
84	Polonium	210	0.80 ~10^{-3} percent

Table 1.--Principal gamma rays in the uranium and thorium series--
Continued.1/

Z	Isotope	A	Energy in mev of gamma rays
			Thorium 232 series
90	Thorium	232	~0.055 (0.075) Coincident with 24 percent of alpha particles
88	Radium	228	~0.03
89	Actinium	228	0.058, 0.129, 0.184, 0.338, 0.462, 0.914, 0.969
90	Thorium	228	0.0843 2 percent e/γ ~12 with 28 percent of alpha particles
88	Radium	224	0.241 e_k/γ0.1 with 4.6 percent of alpha particles
86	Emanation	220	
84	Polonium	216	
82	Lead	212	$\gamma_1$0.115, $\gamma_2$0.176, $\gamma_3$0.238, $\gamma_4$0.249, $\gamma_5$0.299 γ_3 ~40 percent of disintegrations; γ_5 ~4 percent of disintegrations
83	Bismuth	212	With α: 0.040 (~4 percent), 0.144, 0.164, 0.288, 0.328, 0.432, 0.452, 0.472 With β: 2.20, 1.81 (~7 percent), 1.61 (~7 percent), 1.34 (~5 percent), 1.03 (~6 percent), 0.83 (~19 percent), 0.72 (~19 percent)
84	Polonium	212	
81	Thallium	208	(Equilibrium disintegrations of Tl 208 only 35 percent of other elements in series owing to branching of Bi 212) 2.62 (~100 percent, e/γ~0.002); 0.859 (~15 percent, e/γ~0.02); 0.582 (~80 percent, e/γ~0.02); 0.510 (~25 percent, e/γ~0.08); 0.277 (~10 percent, e/γ~0.3)

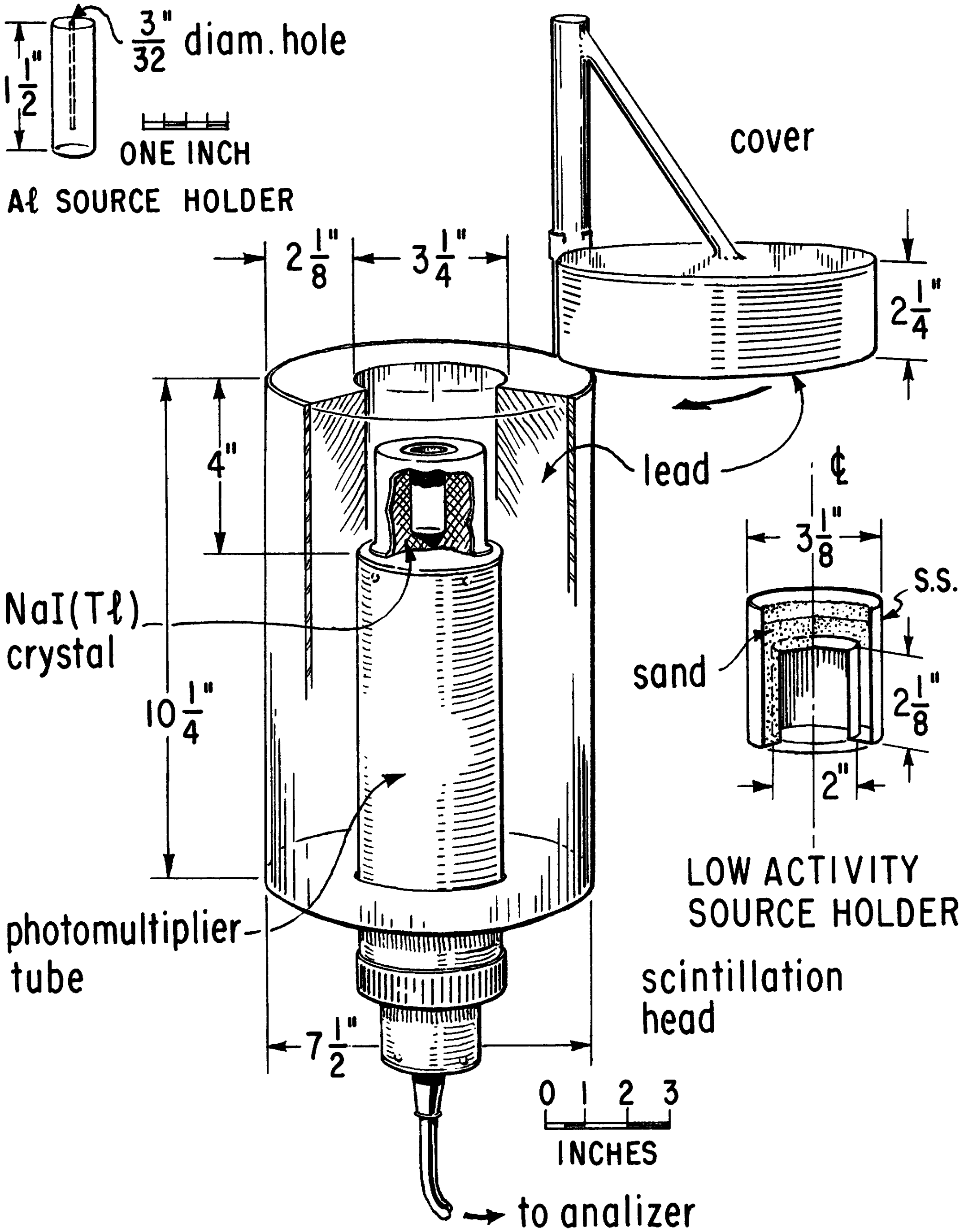
1/ From Hollander, Perlman, and Seaborg (1953). Unless otherwise stated, abundance is given as the number of unconverted gamma rays emitted per 100 disintegrations. The ratio e/γ is the number of conversion electrons emitted relative to the number of unconverted gamma rays emitted.

spectrum at the same time. Both the channel width and the voltage level at which it operates may be calibrated in terms of gamma-ray energy. In the work described here, two channels are used as the analyses depend on the measurement of ratios.

INSTRUMENTATION

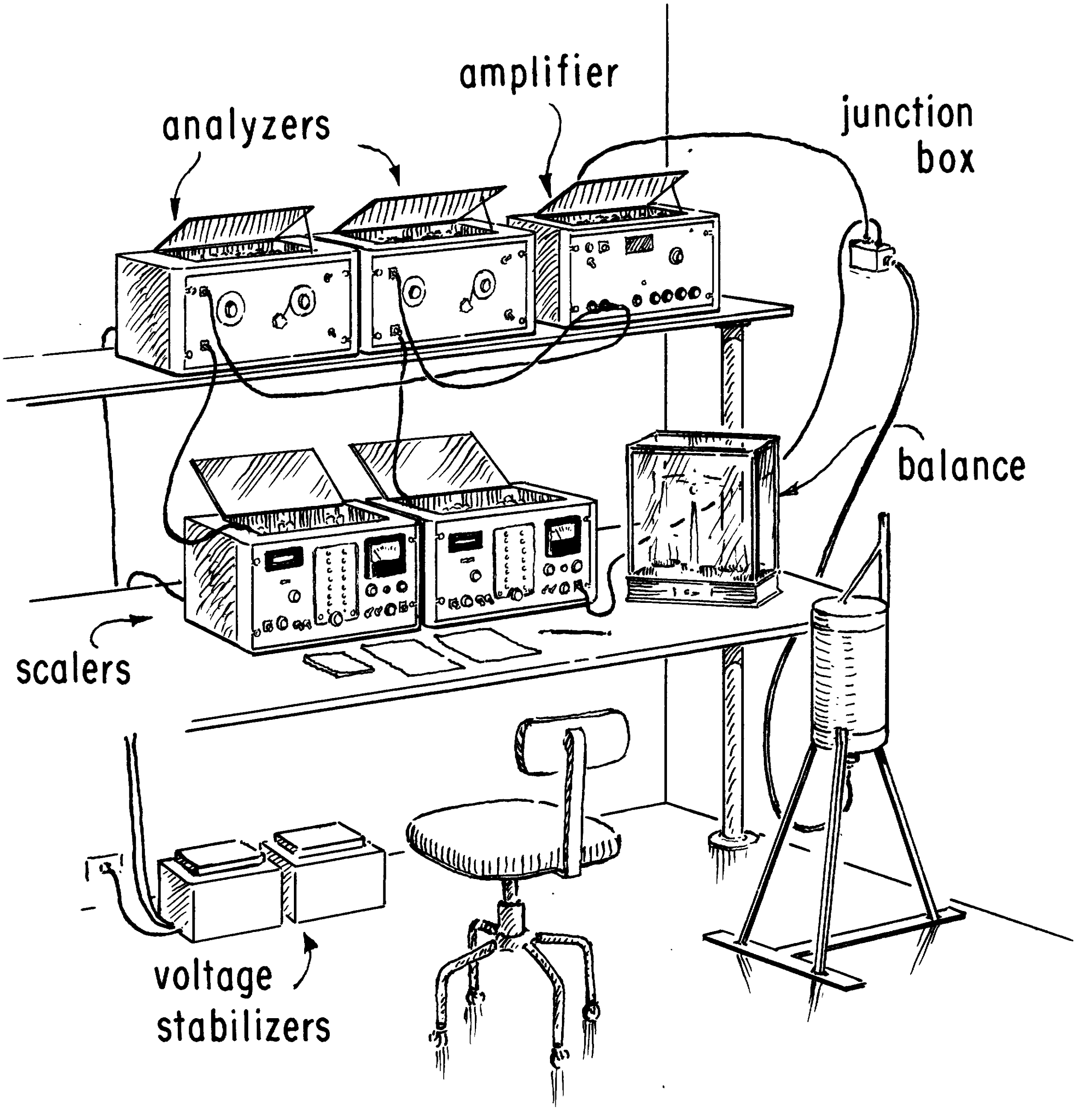
An initial survey of the spectra of the uranium and thorium series showed that the peak due to lead 212 in the thorium series occurred in a region of fairly constant response in the uranium series and might best fulfill the needs for discrimination of the two series. Furthermore, the higher counting rates at the low energy part of the spectrum make it desirable to work in this part of the spectrum if possible.

In a radioassay with gamma rays of this low energy, it is necessary to keep the thickness of the source small or a correction must be made for source absorption if a container of constant dimensions is used. The sample holders for a source in a well-type crystal and for a source surrounding a crystal are illustrated in figures 1 and 2, together with the arrangement of the scintillation head, shield and two-channel analyzer. The source holder for the well-type crystal is an aluminum cylinder 15 mm in diameter with a 3-mm hole drilled in the center to carry the sample; it is used exclusively for the range of sample activity considered in this part of the report. A wall thickness of 6 mm excludes the most energetic beta rays and provides a centered support for the thin column of sample powder. The crystal is thallium-activated sodium iodide, 1 3/4 in. in diameter by 2 in. high, in a light-tight aluminum container. The crystal faces a 5819 photomultiplier tube. The scintillation crystal is housed in a lead shield cast in the form of a cylinder with a wall



DETAIL OF PHOTOMULTIPLIER, CRYSTAL SOURCE HOLDER AND SHIELD

Figure 1



TWO-CHANNEL SCINTILLATION SPECTROMETER

Figure 2

thickness of 2 in. (Irvine, 1954). This has a lead cap mounted on a vertical rod which swings sideways to permit the changing of samples. The pulse signal is amplified linearly and the appropriate bands from the spectrum are admitted by the analyzers and recorded on the scalers. The supply to all electronic components is regulated for voltage, and large variations in temperature are avoided by air conditioning and an open array of the components.

RELATIVE SPECTRA OF THE URANIUM AND THORIUM SERIES

Relative scintillation spectra of the gamma radiation from the uranium and thorium series in secular equilibrium were obtained from analyzed mineral sources containing these elements. The New Brunswick Laboratory of the U. S. Atomic Energy Commission supplied analyzed samples of thorium-free uraninite, and thorium-rich monazite that were used as standards. The monazite was not entirely free from uranium, so that the contribution due to the small amount of uranium present had to be removed from all analyses in order to get an effectively pure thorium standard. The spectrum for the uranium series is shown in figure 3, and that for the thorium series in figure 4. In figure 5 a comparison of thorium and uranium in the range up to 400 kev shows the photopeak due to lead 212 in more detail as it contrasts with the uranium spectrum in this interval.

These spectra were obtained by using a channel width of from 15 to 60 kev and taking separate counts at intervals of 5 to 25 kev over the entire range. The individual points are not shown in the plots because they are so numerous at the low energy end as to obscure the character of the plot, and also because the number of counts in each run was not constant

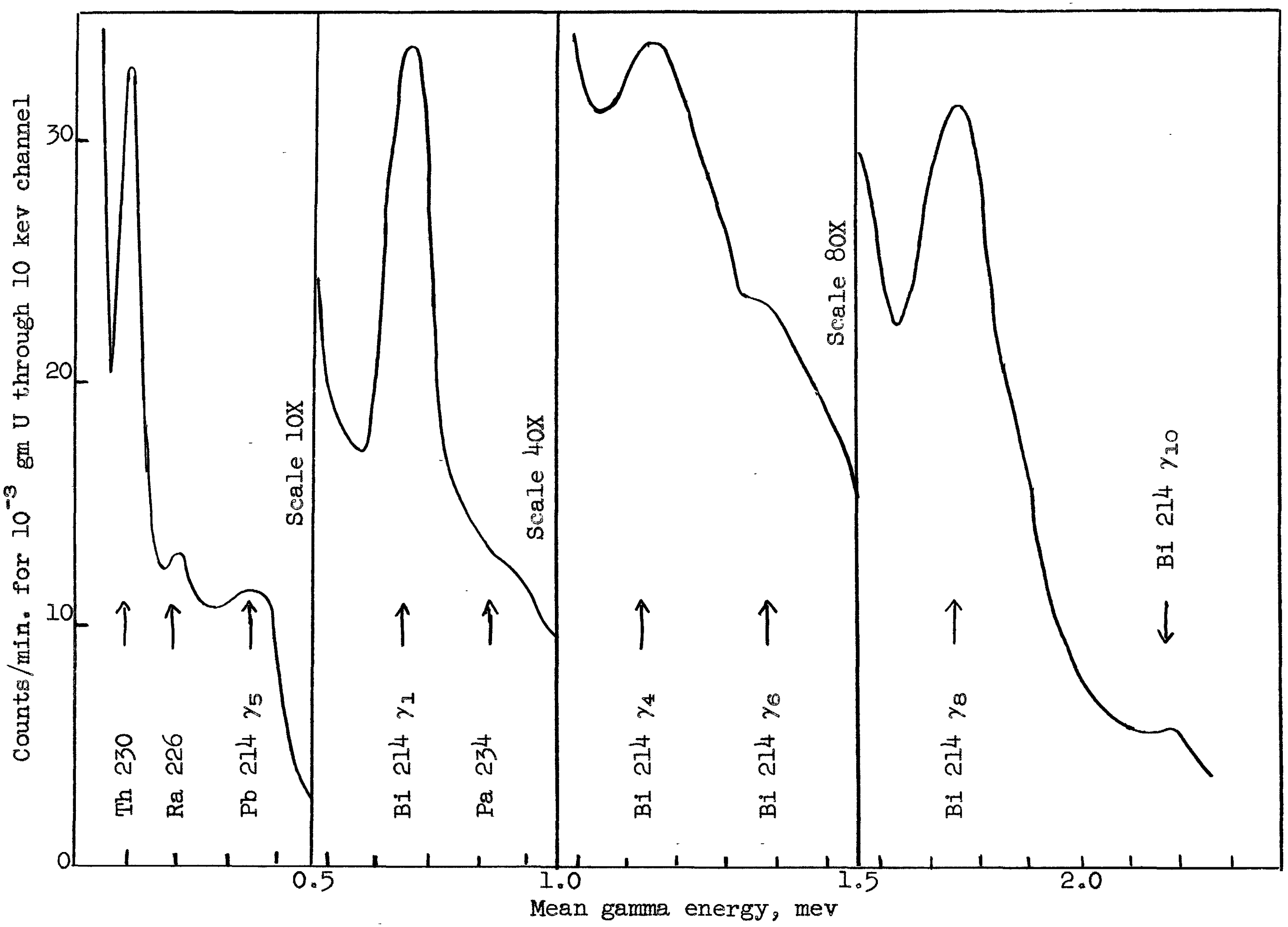


Figure 3.--Gamma scintillation spectrum for uranium series in equilibrium.

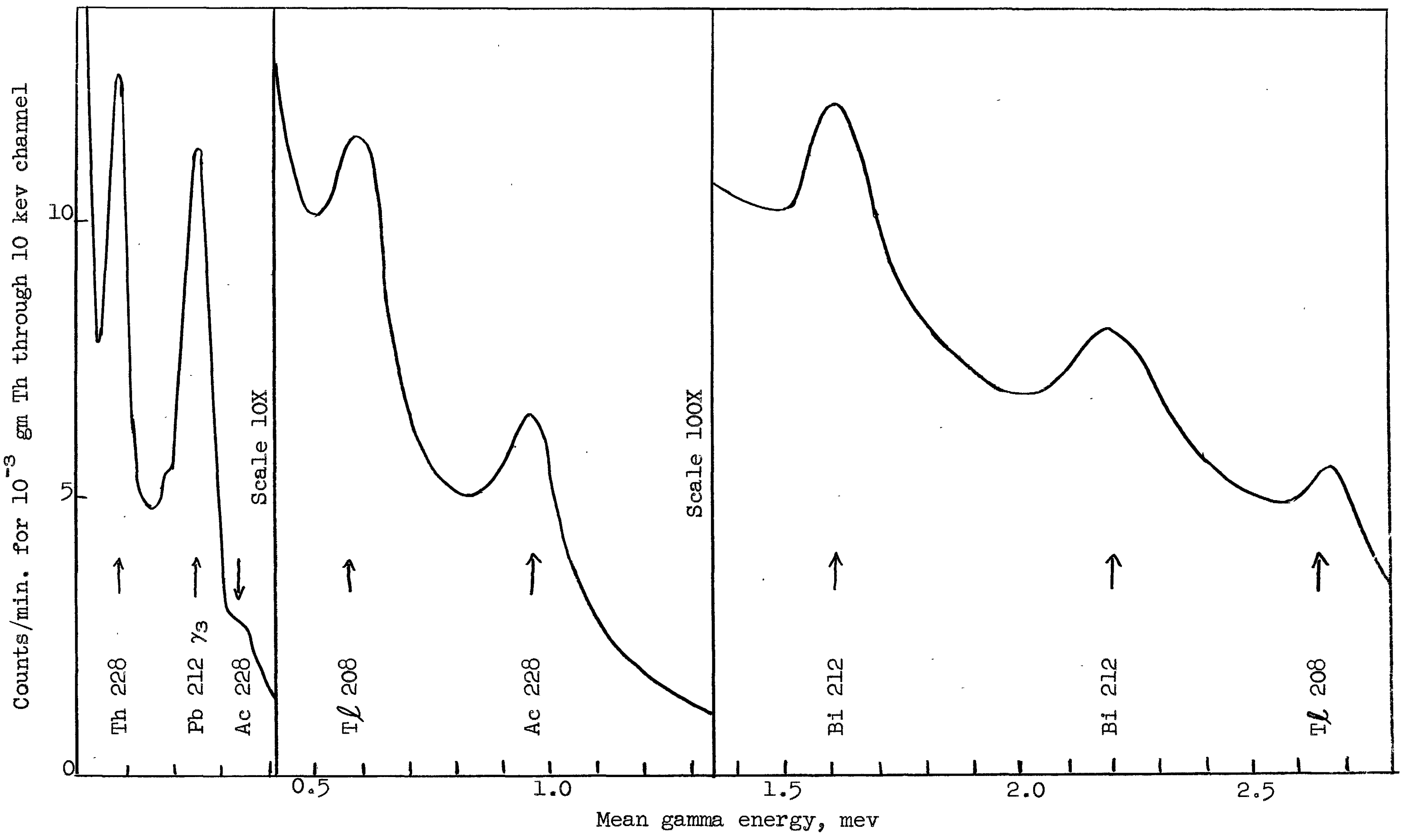


Figure 4.--Gamma scintillation spectrum for thorium series in equilibrium.

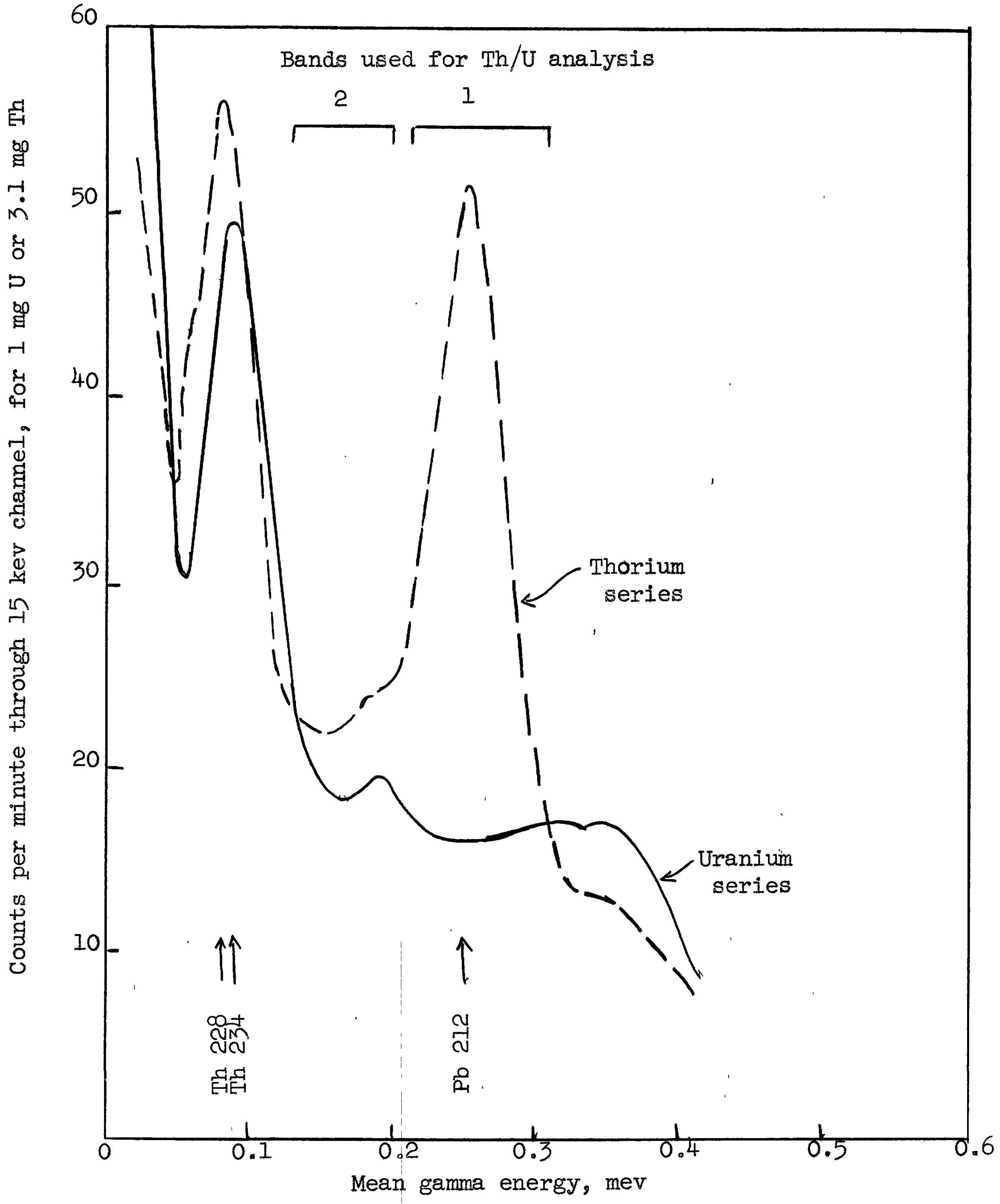


Figure 5.--Comparison of U and Th series spectra below 0.4 mev showing bands used for Th/U analysis (plotted on basis of equal parent disintegrations in each source).

so that the errors differ for each point. Over much of the low-energy parts of the spectra, the standard deviation is less than two line widths. The resolution was diminished by the use of the 6-mm aluminum beta absorber over the whole range, although it was not required except in the low energy region.

METHOD OF ANALYSIS AND TESTS

From figure 5 the determination of the ratio of thorium to uranium in a sample depends on the comparison of a count from a band centered at 238 kev with a band centered at 180 kev. The width of the channels used for this ratio measurement is chosen on the basis of highest overall precision per unit of time required for the entire analysis. The factors entering this choice are shown in figure 6. A single analysis includes one or more calibrations with a standard. As these calibrations are made to correct for instrument drift, the drift error depends on the time interval between the calibrations. The counting error is the square root of the total count. The total count per unit time is proportional to channel width, which is inversely proportional to the resolution of the lead 212 peak. It can be seen that the optimum channel width depends on the drift rate of the instrument.

A determination of the plateau width for the lead 212 photopeak versus channel width is also shown in figure 6. This plateau was defined as the width at the top of the peak included in a 3 percent drop from the peak value, the width being given in terms of percent of change of the base line discriminator. This plot has a slope indicating that the sharpness of the peak decreases, and therefore base line drift causes reduced error as channel width is increased. This compensates to some

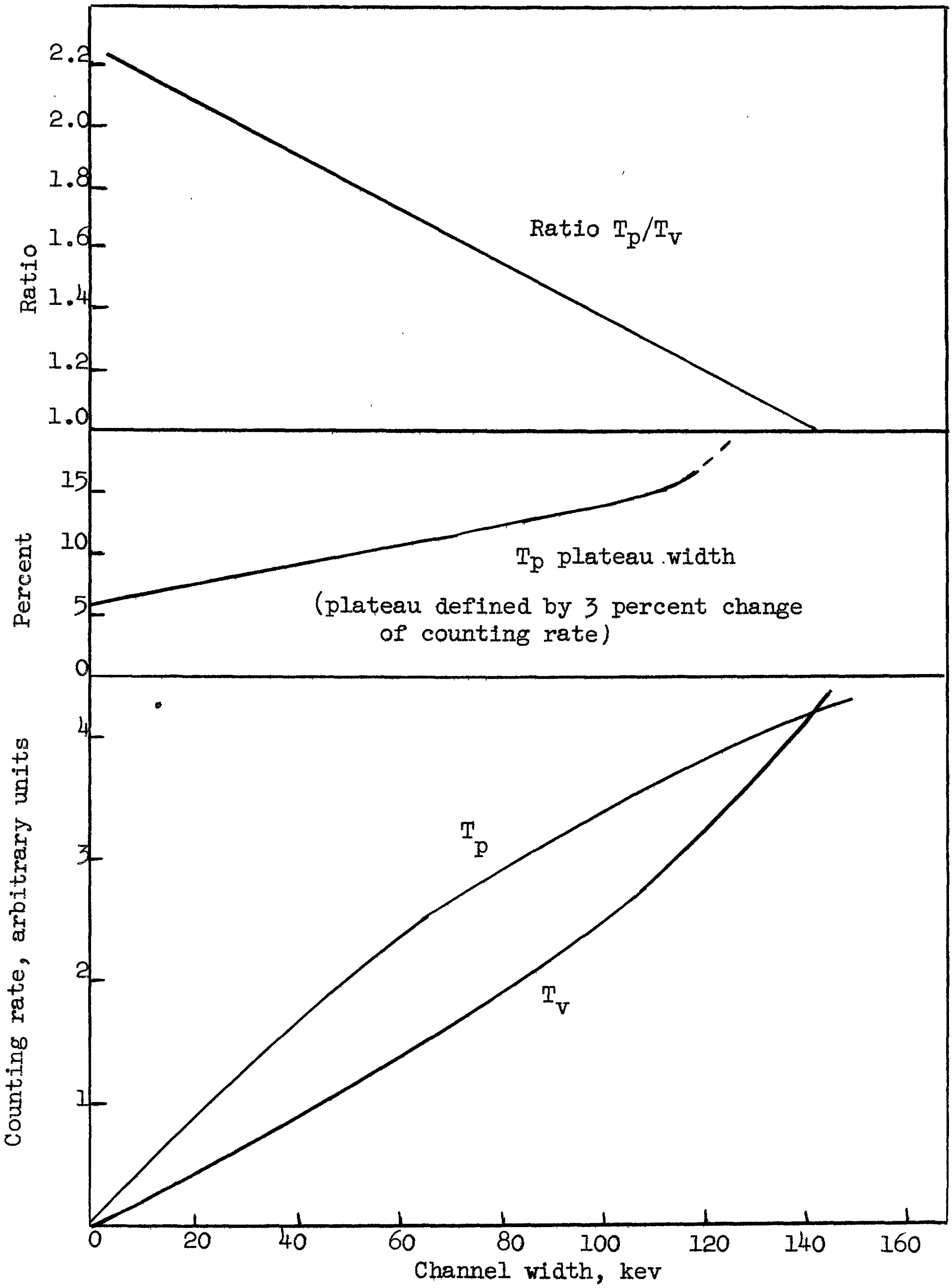


Figure 6.--Factors in selection of optimum channel width for thorium discrimination.

extent the effect due to the square root variation of the counting error and creates an optimum for the channel width which is still dominantly dependent on drift rate.

It is important to maintain a knowledge of the drift rate of each channel. This is done by plotting the calibration values in each channel for a standard source and noting whether the points make a fairly smooth plot. In the present instrument the total amplitude of the drift variation may be as much as 5 percent, but the period of such a variation is generally several days. An operational procedure may be established in which the channel width is left constant and analyses are discarded in which the drift variation during the time of the run exceeds a certain amount.

Operating procedure is as follows. A standard uranium source (3,000 counts per minute is satisfactory) is used to correct for variations in the counting response. After a number of measurements of this standard, a mean value for each channel is found, and some arbitrary value close to the mean value is chosen to represent a datum for the channel. Henceforth, all readings found are corrected according to how much the standard source records above or below this datum at that time. Thus, if the standard source is run before and after an unknown and shows that channel number 1 is counting 5 percent below the datum level, the reading for the unknown is corrected upward by 5 percent to make it homogeneous with the fixed calibration constants, which are also based on the datum values.

When a run is started on a group of samples the two channels are centered on the valley and the peak respectively by using a thorium-rich source and varying the base line discriminator a small amount each way. A standard thorium source is then run, preceded and followed by the standard uranium source which is always used to correct the measured values for instrument drift, as explained above. After this, the unknowns are run alternately with the uranium standard. Calibrations with the standard thorium source are made only when needed; usually the thorium calibration holds well for a number of runs. As only ratios are involved all variations in the background level and in the scintillation head and amplifier cancel out because they are common to both channels.

The calculation of the quantity of uranium and thorium in an unknown is as follows. Counting rate means net counting rate after background has been removed and corrected for drift by calibration with the uranium standard.

U = weight of uranium standard in micrograms.

U_v = mean valley channel counting rate for the uranium standard, i.e., the datum value, in counts/minute.

U_p = ditto for the peak channel.

T = weight of thorium in thorium standard in micrograms.

T_v = valley channel counting rate for the thorium standard corrected for drift and also corrected for the small amount of uranium in this standard in counts/minute.

T_p = ditto for the peak channel.

R_v = valley channel counting rate for unknown sample in counts/minute.

R_p = ditto for the peak channel.

X = counting rate in R_v due to uranium only in counts/minute.

$X \frac{U_p}{U_v}$ = counting rate in R_p due to uranium

$(R_v - X)\frac{T_p}{T_v}$ = counting rate in R_p due to thorium

$$X = R_v \left\{ \frac{\frac{T_p}{T_v} - \frac{R_p}{R_v}}{\frac{T_p}{T_v} - \frac{U_p}{U_v}} \right\}$$

If $\frac{T_p}{T_v} = Q_t$; $\frac{R_p}{R_v} = Q_r$; $\frac{U_p}{U_v} = Q_u$,

Uranium in unknown $= R_v \frac{U}{U_v}\left\{\frac{Q_t - Q_r}{Q_t - Q_u}\right\}$ micrograms.

If $\frac{Q_t - Q_r}{Q_t - Q_u} = F$; $\frac{U}{U_v} = C_u$; $\frac{T}{T_v} = C_t$, then:

Uranium in unknown $= R_vC_uF$ micrograms, and

Thorium in unknown $= R_vC_t(1-F)$ micrograms.

The uranium, thorium, and Th/U ratio are shown as functions of R_v and Q_r in figure 7. With 6 mm of aluminum used for the absorption of beta particles, and 24-kev channel widths, the calibration constants for the instrument and particular source geometry were found to be as follows:

$C_u = 30.5$ micrograms of uranium per count per minute

$Q_u = 0.825$

$C_t = 91$ micrograms of thorium per count per minute

$Q_t = 1.978$

Average background in both channels is about 9 counts per minute. This channel width is used for materials of ore grade; twice this channel width may be used for materials lower in activity to reduce the overall time per analysis as less time is spent for calibration and correction for instrument drift. Samples weighing as much as 0.5 gram may be used in the ore-grade source holder in the crystal well.

A factor of 10 in sample size is gained if the source absorption is taken into account either by correction or by making standards of approximately the same material as the unknown and still using the well-type crystal. Another large factor is gained by using samples of 150 grams surrounding the crystal.

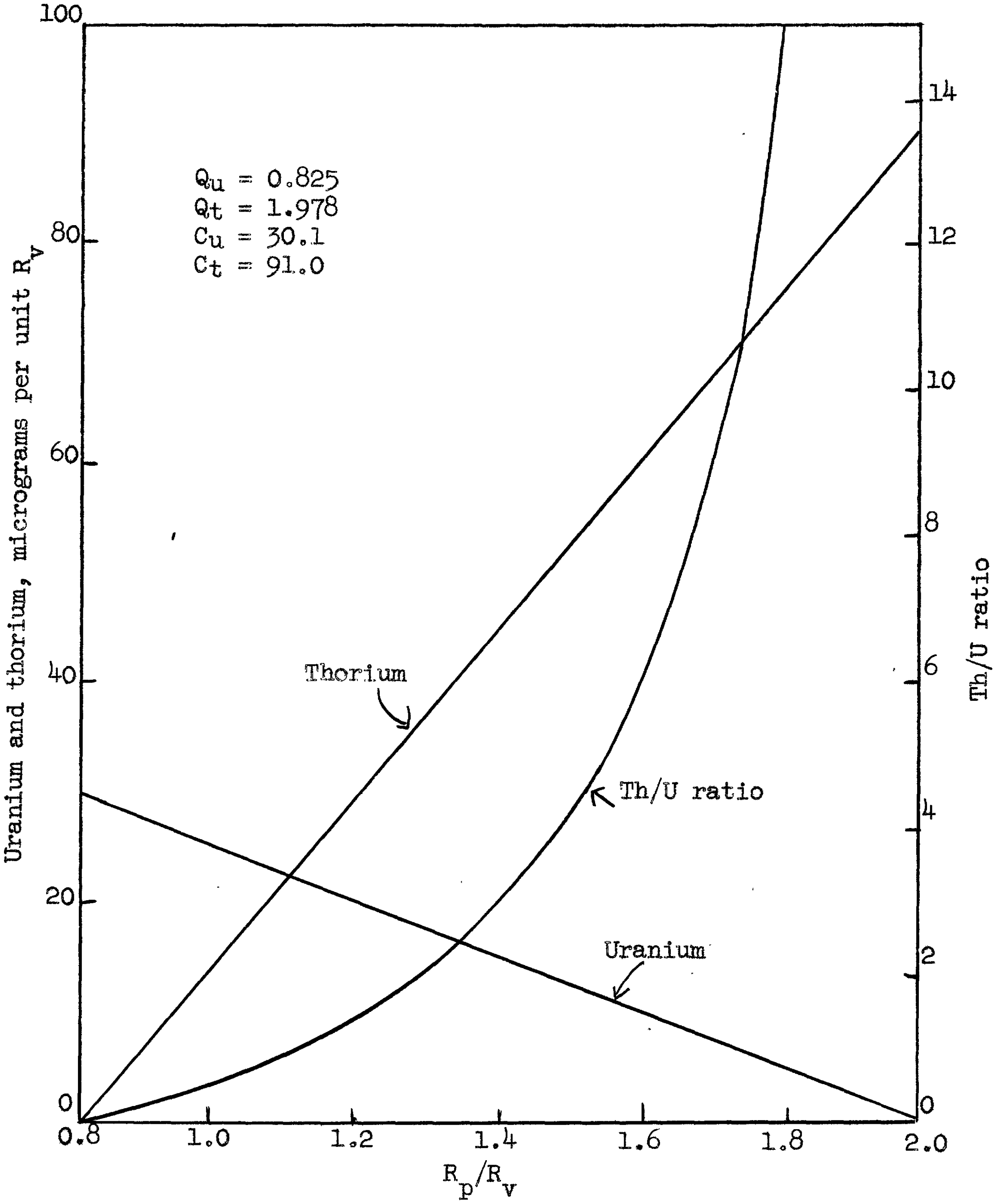


Figure 7.--Uranium, thorium and U/Th as functions of R_v, Q_r.

The precision of the method depends on the relative abundance of the uranium and thorium in the sample. The smaller the amount of thorium relative to uranium, the greater is the precision of the uranium analysis and the less the precision of the thorium analysis, and vice versa. The thoroughness of the drift correction is more important than added counts after a certain number is reached.

Let E_1 and E_2 be the errors as standard deviations in R_v and R_p, which include both counting statistical error and error in drift correction. E_3 and E_4 are similar errors in T_v and T_p. The error E_5 associated with Q_r is

$$E_5 = Q_r\sqrt{\left(\frac{E_1}{R_v}\right)^2 + \left(\frac{E_2}{R_p}\right)^2}$$

and the error E_6 associated with Q_t is

$$E_6 = Q_t\sqrt{\left(\frac{E_3}{T_v}\right)^2 + \left(\frac{E_4}{T_p}\right)^2}$$

The error in the uranium measurement is obtained from

$$(R_v \pm E_1) = \left\{\frac{Q_t \pm E_6 - Q_r \pm E_5}{Q_t \pm E_6 - Q_u \pm 0}\right\}$$

As R_v and R_p are usually about equal, their errors are similarly almost the same, so that

$$E_5 \cong \sqrt{2}\left(\frac{E_1}{R_v}\right)$$

Q_t is always about 2 and $\frac{E_3}{T_v}$ is about equal to $\frac{E_4}{T_p}$ because the drift correction error is largest and common to both. Therefore,

$$E_6 \cong 2\sqrt{2}\left(\frac{E_3}{T_v}\right)$$

Except in unusually bad periods of drift (which can be ruled out) the errors in R_v and R_p are estimated to be about 1 percent standard deviation, and the errors in T_v and T_p about 0.7 percent standard deviation. This gives

$$\text{Uranium} = C_u(R_v \pm 0.01R_v)\left\{\frac{2 \pm 2\sqrt{2}(.007) - Q_r \pm \sqrt{2}(.007)}{2 \pm 2\sqrt{2}(.007) - 0.8 \pm 0}\right\}$$

Q_r varies from 1 to 2 so that F varies between 0 and 1, and the estimated error in the uranium determination varies from 3.3 percent to infinity depending on whether the activity is 100 percent or 0 due to uranium. A plot of estimated errors on the bases outlined above is given in figure 8. The curves for uranium and thorium differ because of the fact that uranium has approximately three times the activity of thorium.

The method and the estimate of precision were tested on a number of samples covering a range of the ratio of uranium to thorium. These test samples were made up of amounts of uranium and thorium standards mixed together in different proportions so that, except for possible inhomogeneity in the standard material, the test samples were of accurately known composition. The results of test analyses are given in table 2. The column listing the total time for the analysis refers to actual time, which for most samples exceeded the time that would be needed if the procedure were made routine and only the necessary and optimum number of counts were made. The errors of most of the analyses are well within the estimated precision given in figure 8. The larger errors occurred at times when the running plot of instrument drift showed unusually large fluctuation. These fluctuations could have been eliminated on the basis of a specified maximum permissible drift rate.

SOURCE GEOMETRY

In reference to the cross section of the source in figure 9,

A_γ = gamma events in unit length of source per unit time, at energy γ

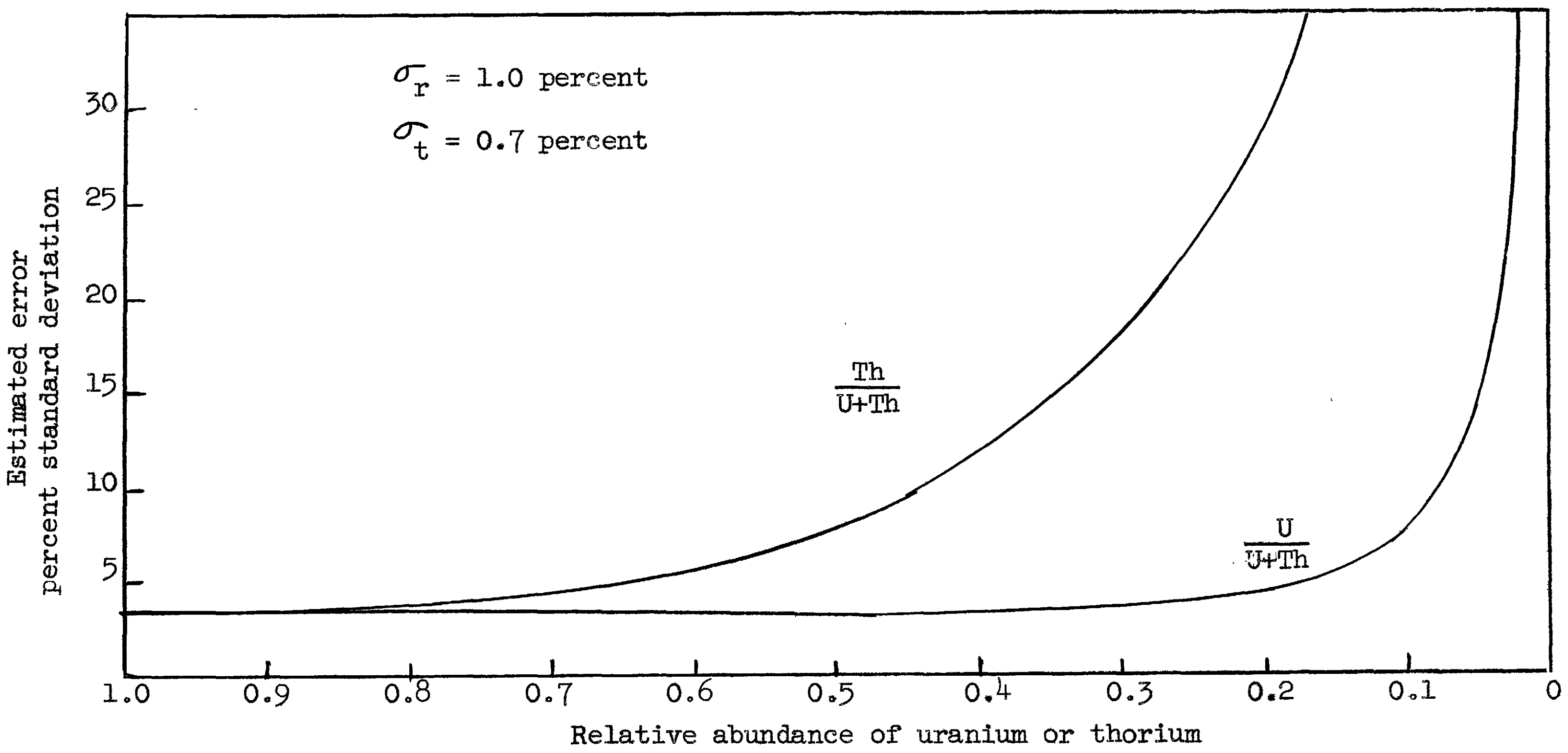


Figure 8.--Estimated precision for different proportions of uranium and thorium, for single comparison against standard.

Table 2.--Results of the method on a number of test samples.

Test sample	Uranium (mg)	Thorium (mg)	Measured uranium (mg)	Percent error	Measured thorium (mg)	Percent error	Total analysis time (min)1/
1	56.3	0.05	56.0	0.5	0	--	30
2	58.2	31.1	56.8	2.4	29.8	4.0	22
			56.8	2.4	31.3	0.6	50
			56.7	2.4	32.5	4.8	45
Mean			56.8	2.4	31.2	0.6	
3	32.7	31.1	33.5	2.6	29.1	6.5	39
			33.3	1.8	30.2	2.9	55
Mean			33.4	2.2	29.7	4.7	
4	24.7	31.3	26.2	6.1	27.6	11.7	45
			25.1	1.6	30.1	3.8	34
			25.1	1.6	30.1	3.8	24
Mean			25.4	3.1	29.3	6.4	
5	15.4	31.5	14.4	6.4	34.0	8.0	42
			15.5	0.6	30.8	2.1	25
Mean			14.9	2.9	32.4	2.9	
6	1.22	32.1	1.22	0.1	31.2	3.0	100

1/ Includes calibrations, weighing, and calculations.

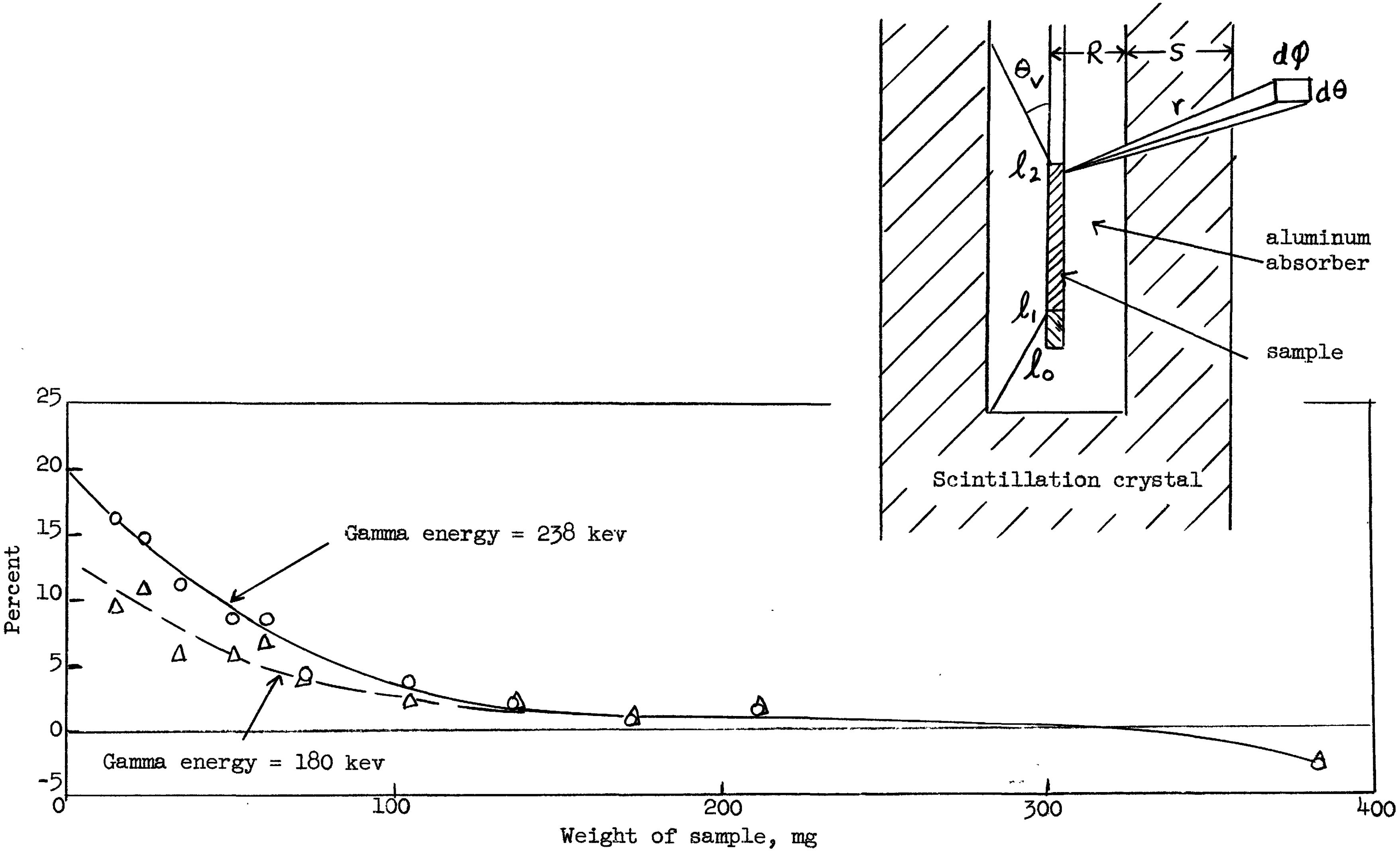


Figure 9.--Deviation from normal value of count per unit sample weight, as function of total sample weight, showing absorber end effect.

μ_a = absorption coefficient of absorber at energy ν

μ_s = photoelectric absorption coefficient of NaI(Tℓ) at energy ν

θ_ν = an angle of incidence above which most of the unabsorbed gamma rays from the region $\ell_1 - \ell_2$ emerge, and below which the proportion of gamma rays is small enough so that the end effects of the absorber cause an error that is either not significant or else may be corrected simply in the flat part of the accompanying curve

K = constant of proportionality which involves the proportion of pulses equivalent to energy ν that result from Compton scattering, efficiency, and other factors.

Any unit of source length in the interval ℓ_1 to ℓ_2 in which there is no significant end effect will give rise to a count of gamma rays at energy ν as follows:

$$C = \frac{A_\nu K}{4\pi r^2} \int_0^{\pi}\int_0^{2\pi} r^2 \sin\theta \exp\left[-(R\mu_a/\sin\theta + S\mu_s/\sin\theta)\right] \cdot d\theta \cdot d\phi$$

$$= A_\nu K \int_0^{\frac{\pi}{2}} \exp\left[-\csc\theta(R\mu_a + S\mu_s)\right] \cdot \sin\theta \cdot d\theta$$

The source length in the interval ℓ_0 to ℓ_1 contributes a count that is greater than any equal length in the interval ℓ_1 to ℓ_2 owing to the end of the absorber. The total count in the interval ℓ_0 to ℓ_1 is

$$C_o = \frac{A_o K \ell_1}{2} \int_0^{\frac{\pi}{2}} \exp\left[-\csc\theta(R\mu_a + S\mu_s)\right] \cdot \sin\theta \cdot d\theta + \frac{C_o}{B(\mu)}$$

where B is some value between 1 and 2 depending on the geometry of the bottom of the hole with respect to the end of the absorber. For any fixed source cylinder B is also a function of μ which varies with the gamma energy being measured.

When the length of the sample in the holder exceeds ℓ_2, the upper end of the crystal affects the count. The two end effects cause a slight decrease of count with increase of sample volume in the region ℓ_1 to ℓ_2

as may be seen in the curves in figure 9. The integrals together with the factor K are evaluated only by direct calibration using increasing amounts of sample, but they serve to explain the shape of the curves and the reason why the effect at the higher energy p is less than the effect at the lower energy ν.

In practice it may be sufficient to keep the sample weight within the limits of the flat part of the curve without correction as was done with the test samples listed in table 2. Another way would be to fill the length ℓ_0 to ℓ_1 with inactive material of about the same composition as the sample, before adding the sample, to remove the lower end effect. For precise work it might be necessary to use such curves as given in figure 9 and to correct values of R_ν, and R_p and Q accordingly.

ACKNOWLEDGMENTS

This research was conducted by the U. S. Geological Survey on behalf of the Division of Research of the U. S. Atomic Energy Commission. The New Brunswick Laboratory of the Commission supplied analyzed samples of thorium-free uraninite and thorium-rich monazite for standards. The instruments necessary for the investigation were made available by the Office of Naval Research, U. S. Department of Defense.

REFERENCES

Eichholz, G. G., Hilborn, J. W., and McMahon, C., 1953, The determination of uranium and thorium in ores: Canadian Jour. Physics, v. 31, p. 613.

Hofstadter, R., and McIntyre, J. A., 1950, Gamma-ray spectroscopy with crystals of NaI(Tl): Nucleonics, v. 7, no. 3, p. 32.

Hollander, J. M., Perlman, I., and Seaborg, G. T., 1953, Tables of isotopes: Rev. Modern Physics, v. 25, p. 469-651.

Irvine, J. W., 1954, Simple efficient shield for well-type scintillation crystals: Nucleonics, v. 12, no. 10, p. 62.

Kahn, B., and Lyon, W. S., 1953, Scintillation spectrometer in radiochemical analysis: Nucleonics, v. 11, no. 11, p. 61.

Pierson, D. H., 1951, Alpha-particle assay and the measurement of the thorium-uranium ratio in radioactive ores: Phys. Soc. Proc., Sec. B, v. 64, p. 876.