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MINES BRANCH

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R 85

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A LABORATORY STUDY OF GAMMA-RAY SPECTRA AT THE SURFACE OF ROCKS

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APRIL 1961

Mines Branch Research Report R 85

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SYNOPSIS

A 100-channel gamma-ray spectrometer was used to investigate the changes in gamma-ray spectra resulting from variations in the thickness of radioactive mineral sources. Uranium, thorium and potassium were studied separately in the form of prepared mineral aggregates and together in the form of large rock specimens.

Rocks are shown to have distinctive gamma-ray spectra that reflect their radioelement content. For common rocks, the intensity of gamma radiation from potassium predominates over that from either uranium or thorium and may exceed their sum.

With improvement of techniques of measurement, the gamma-ray spectrometer should be a useful tool in geological correlation and may make possible simultaneous determination of U, Th and K in rocks "in situ".

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Direction des mines Rapport de recherches R 85

ÉTUDE AU LABORATOIRE DE SPECTRES DE RAYONS GAMMA À LA SURFACE DES ROCHES

par

A.F. Gregory* et J.L. Horwood**

RÉSUMÉ

On a utilisé un spectromètre à rayons gamma de bande 100 pour étudier les modifications des spectres des rayons gamma par suite de variations de l'épaisseur de sources minérales radioactives. L'uranium, le thorium et le potassium ont été étudiés séparément d'abord, sous forme d'agrégats minéraux préparés, puis tous les trois ensemble, sous forme de gros échantillons rocheux.

On constate que les roches ont des spectres de rayons gamma distincts qui reflètent leur teneur en radioéléments. Dans le cas des roches ordinaires, l'intensité de la radiation gamma attribuable au potassium est supérieure à celle de l'uranium ou du thorium et peut même dépasser la somme des deux.

Avec l'amélioration des techniques de mesure, le spectromètre à rayons gamma devrait un instrument utile en matière de corrélation géologique et pourra même permettre de déterminer simultanément les teneurs en U, Th et K des roches en place.

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CONTENTS

Page
Synopsis i
Résuméii
Introduction 1
Apparatus and Methods of Investigation 3
Sources
Technique of Measurement 4
The Nature of a Gamma-Ray Spectrogram
Method of Interpreting the Spectrograms11
Gamma-Ray Spectrograms from Prepared Source Aggregates.13
Results of Experiments
Extrapolation of Results to Generalized Source-Detector Geometries
Approximation of the Effects of Source Density Variations
Gamma-Ray Spectrograms from Rock Specimens
Results of Experiments
Approximate Contribution of Each Radioelement to the Gamma Radiation Field from Rocks
The Possibility of "In Situ" Determination of the Radioelement Content of Rocks
Conclusions43
Acknowledgments45
References
Appendix A - Description of Sources
Appendix B - Instrumentation

ŧ,

٧.,

*

jii

FIGURES

No.		Page
1.	Sketch of source-crystal geometry (vertical section)	. 5
2.	Variation of count-rate with source thickness for selected energy bands - potassium	. 14
3.	Variation of gamma-ray spectrogram with source thickness - potassium	. 15
4.	Variation of count-rate with source thickness for selected energy bands - uranium and thorium	. 16
5.	Variation of gamma-ray spectrogram with source thickness -uranium	. 17
6.	Variation of gamma-ray spectrogram with source thickness - thorium	. 18
7.	Selected count-rate increment curves	. 21
8.	Gamma-ray spectrograms of four rock specimens	. 31
	TABLES	
1.	90% Emission Thicknesses for Source Aggregates Completely Filling the Solid Viewing Angle	. 23
2.	90% Emission Thicknesses for Rock Sources	. 29
3.	Comparative Integral Count-rates from Spectrogram of Standards and St. George Granite (Figures 3, 5, 6, 8)	s . 34
4.	Comparative Count-rates for Selected Energy Lines in the Spectrograms of Standards and St. George Granite (Figures 3, 4, 6, 8)	. 34
5.	Contribution of Radioelements to Rock Activity Measured with Various Counting Thresholds	. 36
6.	Activity Ratios for Unit Amounts of Radioelements	. 38
7.	Comparative Radioelement Analyses of Four Rocks .	. 42

INTRODUCTION

Rock and soils at the surface of the earth contain natural radioactive isotopes, which emit alpha, beta and gamma radiations. All three radiations may be of interest in laboratory investigations, but only gamma rays penetrate rock and air sufficiently to allow practical geophysical measurements in the field. Of the more than thirty gamma-ray emitting isotopes which occur in nature, only radio-potassium (K-40) and members of the uranium and thorium series are important geological sources of gamma radiation. The isotopic abundances, probabilities of geological concentration and gamma-ray energies of the other natural emitters are so low that these isotopes may be neglected in radiometric surveys.

To date, most geophysical measurements of radioactivity have been made with Geiger-Muller counters or scintillation counters, both of which integrate the counting rate over the range of energies accepted by the detector circuit. Differentiation of the energy lines in the spectrum by means of a gamma-ray spectrometer is a logical improvement in the technique of radiometric surveying.

Mero (1) has recently discussed the uses of the gammaray spectrometer in mineral exploration. However, few investigations of the gamma-ray spectra of rocks appear to have been made. The only published gamma-ray spectrograms for rocks which are known

-1-

to the authors are: several used for selecting counting thresholds in radiation prospecting by the energy discrimination technique (2); one illustrating spectral gamma-ray logging in drill holes (3); and two calculated from theory (4, 5).

To investigate the feasibility of geophysical surveying with a gamma-ray spectrometer, a series of experiments was designed to study the variations of spectra which occur with large variations in source thickness and with large variations in air distance between source and detector. Measurements were made under static conditions with several sources of known size and activity. This report is concerned with the first phase of this investigation and describes a laboratory study of the variation in the gamma-ray spectrum which accompanies a progressive increase in source thickness from one-half inch to effective infinity. Radiations from K-40 and members of the uranium and thorium series in equilibrium were studied as separate spectra at the surface of prepared granular aggregates and as composite spectra at the surface of large rock specimens. While the present research was directed primarily toward solving problems in aeroradiometry, the results may also be of interest to other investigators concerned with the measurement of gamma radiation from rocks and soils.

- 2 -

APPARATUS AND METHODS OF INVESTIGATION

Radioactive mineral sources emit gamma radiation with a quantum distribution and intensity reflecting their content of natural uranium, thorium and potassium. This radiation may be analyzed with a scintillation spectrometer (6, 7, 8). The actual gamma-ray emitters and their individual spectra are known, provided the uranium and thorium of both specimen and calibration source are in the same state of equilibrium (usually complete secular equilibrium).

In the present investigation, spectra were measured with a lead-shielded scintillation crystal and ancillary apparatus, as briefly described in the following paragraphs.

Sources

The prepared sources each comprised about 400 lb of radioactive mineral aggregates, which for convenience in handling were placed in thin wooden boxes^{*}. These sources were made up of potash feldspar and mixtures of silica sand with pitchblende and thorite. Several large rock samples were also used. In addition, bags of pitchblende and thorite concentrates were used in a related experiment outside the laboratory. The sources are further described in Appendix A (page 48).

The attenuating effect of the total thickness of wood involved in the thickest source was experimentally determined to be negligible.

- 3 -

Technique of Measurement

The gamma-ray detector was a crystal of NaI(T1), 2 inches thick and 2 inches in diameter, mounted on a photomultiplier tube. The detector was located centrally and 5 3/4 inches below the four-inch-square aperture of a lead castle (Fig. 1). The pulses from the detector were amplified and then sorted by amplitude in a 100-channel, pulse-height analyzer. The amplitude spectra were plotted graphically and were related to an energy scale by calibration with known isotopes. Resolution * of the spectrometer was 10% for the 0.662 MeV photopeak of Cs-137. More detailed information concerning the instrumentation is contained in Appendix B (page 51).

3

In the laboratory measurements, the sources were centred over the aperture ^{**} of the castle. The prepared sources were stacked vertically to various thicknesses of from $\frac{1}{2}$ inch to 48 inches.

It was important to evaluate the optimum cross-sectional area of source which could be used in this investigation. Varying the area of source has only a small effect on the intensity or spectrum of the radiation, provided that such area is larger than

- 4 -

^{*} Resolution is the width of the photopeak at half height, expressed as a percentage of the photopeak energy. It is a measure of the ability of the spectrometer to distinguish between adjacent photopeaks of the spectrogram. Resolution is energy-dependent and thus is normally determined for a standard source, Cs-137 (E = 0.662 MeV).

^{**} The sources were placed above the detector because the method of mounting the crystal required that it be kept upright on the photomultiplier tube.



FIGURE I-SKETCH OF SOURCE-CRYSTAL GEOMETRY Vertical Section

- 5 -

the castle aperture (see Fig. 1). There will be a small component of scattered radiation transmitted through the source from material beyond the solid angle viewed by the detector. The amount of such scattered radiation will vary with the specific activity, size and density of the source and with the energy of its radiation. A large source area obviously approximates natural sources most closely, but requires a very large volume of homogeneous material. However, for the given geometry of the experiment and for sources representing average rocks, it was estimated that practical measurements could be made with small source areas. With such low concentrations of radioelements, the contribution of the radiation scattered from beyond the solid angle of view is almost negligible. The optimum source area was then evaluated by preliminary experiments in which the radiation from potash feldspar ($E_{\gamma} = 1.46$ MeV) was considered to represent the activity and energy of radiation from average rocks. These experiments showed that, within the statistical error of the measurement, and for the geometry of the detector, the spectrum from a source area of one square foot would be equivalent, in both total intensity and energy distribution, to that from a source of nine square feet. For large source thicknesses, however, a source area of one square foot does not fill the solid angle seen by the detector (see Fig. 1). Subsequent measurements (see page 23) suggested that the resulting error in intensity was less than 15% and that this occurred mainly in the low energy portion of the spectrum.

- 6 -

Accordingly, a source area twelve inches square was selected for this investigation. Such sources are considered to be equivalent in cross-sectional area to naturally occurring sources of similar composition, provided correction is made for incomplete filling of the solid angle seen by the detector.

Because of their appreciable thicknesses, the sources acted as shielding of the detector against cosmic and other background radiations. Therefore, inert material (silica sand) was added above the sources to maintain a constant total thickness of material over the detector. Thus, using only inert material with this constant total thickness, background was determined as required and subtracted from the gross source count-rate to obtain the net count-rate for each source thickness.

Counting times were selected to provide good statistics and recordings that could be readily interpreted. The counting time for the uranium and thorium aggregates was 30-40 minutes and for potassium aggregate 50 minutes. Comparable backgrounds were also obtained. For the rock specimens, the counting times were 60, 240, 960 and 3840 minutes. The shortest times provided useful results for all but the rocks of weakest radioactivity.

For some experimental results, it was necessary to make corrections for variation in background and for drift in pulse height (energy calibration). The channel-by-channel reduction of the experimental results and the application of necessary corrections was tedious and time consuming.

- 7 -

THE NATURE OF A GAMMA-RAY SPECTROGRAM

A gamma photon that is absorbed in a scintillation crystal releases energy as a flash of light. The flash is viewed by a photomultiplier tube which generates an electrical pulse that is proportional to the intensity of the light flash and thus proportional to the energy released in the crystal. Such pulses are amplified and sorted according to amplitude into the channels of a pulseheight analyzer. Pulses in each channel are counted separately. In the instrument used, the pulse amplitude is approximately proportional to the gamma-ray energy over the required energy range. A curve showing the number of pulses of a certain amplitude versus the amplitude (usually expressed as gamma-ray energy in MeV) provides information about the quantum distribution of the absorbed radiation.

Gamma-rays may interact with the crystal by three principal processes: photoelectric effect, pair production, and Compton scattering. In the photoelectric effect the photon energy is wholly absorbed and re-emitted within the crystal, so that the amplitude and count-rate of the pulses in the pulse-height distribution are direct measurements of energy and intensity of that photon flux. If the photon energy (E) is greater than 1.02 MeV, pair production may occur, i.e. a positron-electron pair may be created. Annihilation of the positron with a nearby electron results in two

- 8 --

gamma photons of 0.51 MeV energy, which may escape the crystal or may be absorbed in it. Pair peaks may thus appear at locations representing energies of (E-0.51) MeV and (E-1.02) MeV. In Compton scattering, the photon gives up part of its energy through collision with an electron but proceeds with a diminished energy that is dependent upon the angle of deflection from its original path. The photon may subsequently be scattered several times and may escape the crystal. Because of the varying angle of scatter and the many possible interactions, the resulting electrons may have a wide range of energies. Accordingly, the Compton pulse-height distribution is a continuum, the size and shape of which are functions of the quantum energy and the conditions of measurement. With high count-rates and large crystals, summation peaks may also result from the simultaneous absorption of two photons.

Thus the pulse-height distribution measured with a scintillation spectrometer is not a true representation of the gamma spectrum of the incident radiation, i.e., one or more lines corresponding to the discrete quantum energies of the radiation. Rather, the observed pulse-height distribution represents the response of the detector to the incident radiation, i.e., the spectrum of secondary electrons arising from gamma photon interactions with the sodium iodide crystal. In addition, scattered radiation arising from interactions with surrounding material (e.g. lead shielding and phototube) is also recorded. Further, peaks in the pulse-height

- 9 -

distribution may occasionally become flattened and broadened because of (1) statistical variations in the production of pulses, and (2) variations in gain of the photomultiplier tube during the time of measurement. The latter variations may also effect a shift in channel location relative to the energy calibration scale. Thus, the pulse-height distribution, which commonly is referred to as a "spectrum", is here called a <u>spectrogram</u> in order to preclude mistakes in identity with the related, but different, energy spectrum of the investigated radiation. Present techniques do not permit the calculation of the true gamma-ray spectrum from the observed pulse-height distributions (9).

The major features of the gamma-ray spectrogram are: (a) the photopeaks, which represent the major energy lines in the spectrum of the incident radiation; and (b) the Compton continuum, which represents the scattered photons, both in the incident spectrum and in the radiation complex produced by that spectrum's interactions with the crystal and adjacent materials.

Photopeaks provide most of the information concerning the radiation spectrum. Pair, back-scatter and summation peaks may also occur; however, such features were not used as they are more difficult to interpret.

- 10 -

METHOD OF INTERPRETING THE SPECTROGRAMS

Net count-rates and net spectrograms for the various sources were calculated by subtracting backgrounds from gross measurements.

The geometry of the detector and source face remained constant throughout the experiment. Thus the differences observed in the spectrograms for various thicknesses of source were attributed to variations in the emission spectra resulting from the change in source thickness.

The aggregate sources each have separate spectra representing uranium and thorium (both in equilibrium) and potassium. However, the rock specimens emit composite spectra comprising all three of these spectra superimposed and in proportion to the concentration of the radioelements in the rock. Thus for quantitative analysis, the contribution of each of these spectra must be separated, or "stripped", from the rock spectrogram. Because of the complex origin of the rock spectrum and the abundance of Compton-scattered photons in it, the spectrogram for any rock shows less clarity and detail than do spectrograms for component radioelements alone.

The simple spectrogram of gamma radiation from potassium in the feldspar aggregate was studied initially, so that a technique of interpretation could be developed and be subsequently

- 11 -

applied to the more complex spectrograms of the other sources.

The obvious means of interpretation was to relate the height of the photopeak to the pertinent source thickness. However, at an early stage in the investigation it was observed that the count-rate for the particular channel in which a photopeak occurred did not characterize the intensity variations as reliably as did the "area under the photopeak" (i.e. the summation of count-rates for all channels under the photopeak curve). This was attributed to the fact that shifts in channel location relative to the energy calibration and statistical variations in count-rate affected the count in a single channel more significantly than they did the total count from all the channels under the photopeak curve. Accordingly, in this investigation the areas under the spectral curve were determined for selected energy bands, the limits of which depended on the particular spectra being studied. Bands representing recognizable photopeaks were of major interest, but intervening bands and the total for all bands were also measured.

- 12 -

GAMMA-RAY SPECTROGRAMS FROM PREPARED SOURCE AGGREGATES

Results of Experiments

A counting threshold of 600 keV was used for the thick potassium-bearing sources, because there was little spectral detail for the region 200 to 600 keV (see Fig. 3, spectrogram for source thickness of 6 inches measured with a threshold of 200 keV). The counting threshold for the thick uranium-bearing sources was set at 200 keV and for the thick thorium-bearing sources at 150 keV.

Net count-rates for selected bands have been plotted against thickness of source material in Fig. 2 (potassium) and Fig. 4 (uranium and thorium). Other bands, which are not represented in these figures, indicated similar trends. The net spectrograms for source thicknesses of $\frac{1}{2}$, 1, 2, 6, 12 and 18 inches are shown in Fig. 3 (potassium), Fig. 5 (uranium) and Fig. 6 (thorium). The lower energy limit of the spectrograms is shown at 200 keV, since the spectral detail for lower energies was not considered significant in this investigation.

These measurements show that the variation in count-rate with source thickness rapidly approaches a maximum value. The 90% emission thickness^{*} lies between 8 and 12 inches for the prepared source aggregates (Figs. 2 and 4). Material at deeper

^{*} The 90% emission thickness is that thickness of source which emits 90% of the radiation that would be emitted by an infinite thickness of the same source material.





FIGURE 3-VARIATION OF GAMMA-RAY SPECTROGRAM WITH SOURCE THICKNESS-POTASSIUM

Source : Feldspar Aggregate 12.4 % K₂O Observed ------Extrapolated ------

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Source Distance : 6.0 Inches Aperture : 4 Inches Square Channel Width = 14 keV É

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levels does not contribute effectively to the observed source strength. The major change in the shape of the spectrogram on going to greater source thickness is a steady increase in counts in the Compton continuum (Figs. 3, 5 and 6). This build-up of secondary photons results from the scattering in the source material of primary photons originating at depth. Much of this build-up occurs in the first few inches of source thickness. For greater thicknesses the production of secondary Compton photons appears to be partly balanced by increased absorption, and the photopeaks are never completely overwhelmed. A more-or-less constant energy distribution may be attained for large source thicknesses. The spectrograms suggest that much of the scattered radiation comprises photons with an energy less than 1.3 MeV. This is in agreement with theoretical evaluations (10).

Extrapolation of Results to Generalized Source-Detector Geometries

The results obtained from these measurements are peculiar to the source-detector geometry of the experiment. Extrapolation to more generalized geometries approximating naturally-occurring sources is of interest.

The effect of source geometry is apparent in the flattening of the yield curves (Figs. 2 and 4) at thicknesses greater than 8 inches. This flattening is more marked than is to be expected theoretically for sources which fill the solid angle seen by the

-19 -

detector. It was assumed that the flattening at these greater thicknesses resulted mainly from a source configuration that did not completely fill the solid angle. The validity of this assumption is indicated by plotting a count-rate increment curve showing the count-rate from each inch of source thickness. Four such curves representing the major photopeaks for uranium, thorium and potassium are presented in Fig. 7. Note that the plots for all sources break away from a simple exponential relationship (straight line) at a source thickness of between 8 and 9 inches. This is approximately the thickness at which the source configuration ceases to fill the solid angle seen by the detector (see Fig. 1)^{*}. Count-rate increment curves for other bands, not shown in the figure, show similar trends.

The most significant increment curves are those for the bands that represent the highest energy photopeak in the spectrum. Essentially, these curves represent measurements made under conditions of "good" geometry, i.e. primary (unscattered) photons originating from source material within the solid viewing

Further verification of this interpretation of the count-rate increment variation was obtained in a separate measurement using a layer of uranium aggregate, one inch thick and one foot square in area. Count-rate versus distance from crystal was measured for the same source-detector geometry. The effective absorption coefficient of the radiation in the source aggregate was similarly measured. The count-rates were recalculated to an increment curve. This synthesized curve corresponded closely to the pertinent measured count-rate increment curve for uranium, and especially so over the initial straight line portion. Here, also, the divergence from the straight line occurred at about $8\frac{1}{2}$ inches.



-10

angle of the detector. (Actually, as each band of interest has a finite energy range, secondary photons scattered through very small angles are also measured, but numerically these are of minor significance.) Such primary photon data are independent of source material beyond the solid angle of view.

Results for bands of lower energy in the spectrum will include the effects of more abundant scattered photons and thus may represent photons originating in material beyond, as well as within, the solid angle of view. Photons scattered through large angles lose much of their energy (50% to 75% for 0.5 to 3.0 MeV photons scattered through 90°). The spectrograms show that the first few inches of source thickness are responsible for the major portion of the build-up of scattered photons in the Compton continuum. Thus the effect of material beyond the solid viewing angle will primarily be an increase in yield of scattered photons. A part of this effect was evaluated in these measurements for an extension of the source volume up to 4 inches beyond the solid angle of view for source thicknesses less than 8 inches. As mentioned previously (see page 6), the unevaluated secondary photons from beyond the solid viewing angle are of minor significance (i.e., sources of larger area did not appreciably change the appearance of spectrogram).

The count-rate increment curves measured for the first 8 inches of source thickness were extrapolated to greater source thicknesses for complete filling of the solid viewing angle (Fig. 7). The corresponding extrapolated counts for sources filling the solid viewing angle are shown in Fig. 2 (potassium) and Fig. 4 (uranium and thorium). The difference between the extrapolated count and the measured count for source thicknesses over 8 inches is about 10-15% at the greater thicknesses.

For the source materials used in this investigation, the 90% emission thicknesses were calculated. These are tabulated in Table 1 for the energy bands representing the major high-energy photopeak and the full energy range investigated in each spectrum. The true 90% emission thickness for the full energy range investigated in each spectrum probably is only slightly greater than the listed minimum value, for the error due to unevaluated scattered photon flux has been found to be negligible.

TABLE 1

90% Emission Thicknesses for Source Aggregates Completely Filling the Solid Viewing Angle

Source Aggregate	Energy Band	90% Emission Thickness
Potassium (S.G.=1.6)	''1.46 MeV'' (1356 - 1566 keV)	ll inches
	740 - 1580 keV	minimum of 12 inches
Uranium (S.G.=1.5)	''1.76 MeV'' (1700 - 1860 keV)	13.5 inches
	210 - 2290 keV)	minimum of 11 inches
Thorium (S.G. = 1.4)	"2.62 MeV" (2480 ~ 2810 keV)	17 inches
	150 - 2810 keV	minimum of 15 inches

Dixon (11) has presented a method for evaluating the self-absorption in a linear source element. This may be compared with the present measurements of "unscattered" radiation (i.e. the "1.46 MeV" band for potassium, the "1.76 MeV" band for uranium and the "2.62 MeV" band for thorium). Using attenuation coefficients pertinent to the source material (i.e. those of silicon or oxygen), the calculation of 90% emission thicknesses gives 11.0 inches for 1.46 MeV photons ($\mu = 0.0517 \text{ cm}^2/\text{g}$), 13.0 inches for 1.76 MeV photons ($\mu = 0.0467 \text{ cm}^2/\text{g}$) and 16.5 inches for 2.62 MeV photons ($\mu = 0.0390 \text{ cm}^2/\text{g}$). These calculated thicknesses agree favourably with the above experimental determinations.

Bags of thorite and pitchblende concentrates were used in a related series of experiments, and a more generalized evaluation of the thick-source effect was made for these materials under the conditions of relatively small exposed source areas (approximately two square feet) at a relatively large source distance of 22 feet in air from an unshielded scintillation crystal **. The proportion of the solid angle of view occupied by the source was thus effectively constant and was independent of the source thickness. As the bags used in the measurement had appreciable thickness (approximately 9 inches), the shape of the curve could not be as precisely defined as in the laboratory investigation. However, the results for both

*See Description of Sources, page 48.
** The radiation was emitted primarily from the surface facing the detector, as lateral and back scattered emission was restricted by a shield of concrete blocks and steel plates.

- 24 -

energy spectrum and integral count agree well with those presented above and for this reason are not included here. The present integral count results also agree with results measured for pitchblende concentrates with a larger surface area (approximately 17 square feet) at a distance of 150 feet in air from an unshielded crystal (12).

These experimental results all show that the first 12 to 18 inches of source aggregate (S.G. = 1.5) emit 90% of the gamma radiation of an infinitely thick source of similar material under the same conditions of measurement. The precise value of the 90% emission thickness depends upon the energy spectrum of the measured radiation (see Table 1).

The effect of varying the solid angle seen by the detector is difficult to evaluate. If the source fills the solid angle of view and the surface of the source is a constant distance from the detector, then the spectrogram will not be altered, although the count-rate will increase in direct proportion to the increase in solid angle. However, if the source is a flat surface (as in ideal geological sources), an increase in solid viewing angle is attended by variation in the distance from detector to various parts of the source face. It is probable that the major effect will be an increase in the number of photons of all energies with little significant variation in the quantum distribution. The actual variation in count-rate will not be directly proportional to the solid angle of view and will also depend upon the particular detector used. Such variations should not appreciably alter the shape of the count-rate versus thickness curves.

Approximation of the Effects of Source Density Variations

Assuming homogeneity of the distribution of radioelements in the sample of rock, the major extrapolation required to refer the experimental results to geological sources is the allowance for density contrast between the prepared aggregate and the rock.

For elements of low atomic number (< 20) and gamma photons of low energy (0.2 to 3.0 MeV), such as are encountered in measurements of rock radioactivity, the attenuation is predominantly due to Compton scattering. Under these conditions, the mass absorption coefficient is effectively a constant for any one energy, i.e., it is independent of the nature of the absorber. The corresponding linear absorption coefficient, however, is a function of the density of the absorbing medium (i.e. the source material). The source density is dependent on the proportions of the predominant elements in the rock and on the degree of packing of the mineral grains.

Geological sources comprise silicon and oxygen, with lesser amounts of aluminum, iron, calcium, sodium, potassium and magnesium and trace amounts of other elements. In rocks, density contrasts resulting from variation in the elemental composition alone will be relatively small (e.g., densities vary from 2.6 to 3.0 g/cm³ for common acidic to basic igneous rocks), but will result in a small variation in the linear absorption coefficient. However, if a portion of the source volume is occupied by air space, as in a porous rock or an aggregate of mineral particles, density contrasts may be larger (e.g. 1.2 to 2.0 g/cm³ for unconsolidated sediments and 1.7 to 2.7 g/cm³ for sedimentary rocks in various stages of consolidation). An orebody in which high-density uranium or thorium is concentrated relative to the surrounding rock, has a bulk density usually less than 3.0 g/cm³, but locally it may be greater.

The experimental results for the source aggregates may be referred to geological sources, for the same source-detector geometry, by calculating the thickness of rock corresponding to the thickness of aggregate, assuming constancy in the properties of the constituent minerals. As the air absorption in the voids of the aggregate is negligible, the amount of absorbing material remains constant and the scattering and absorption characteristics of the mineral matter will not be changed. Accordingly, for equivalent attenuation, the thickness of rock "in situ" is related to the thickness of source aggregate by the inverse of the ratio of their densities or specific gravities, i.e. $\left(\frac{t_a}{t_r} = \frac{\rho_r}{\rho_a}\right)$. However, as the source material is radioactive itself, there will be a variation in radiation intensity due to changes in geometry. The bottom layer of the rock that contributes radiation will be closer to the detector than will the comparable layer in the aggregate.

The source aggregates may be considered to have an average specific gravity of 1.5. A useful average specific gravity

- 27 -

for common consolidated rocks is 2.7. Thus $t_r = 0.55 t_a$ and radiation from a lamina 14 inches from the surface of the aggregate would undergo the same attenuation as that from a lamina 7.7 inches from the rock surface. The increase in intensity due to change in geometry is not readily calculated from theory and it will vary with the air distance from the source to the detector. Thus, if the air distance is small relative to the thickness of the source, the effect may be large, whereas if this distance is relatively large the effect becomes negligible. For the conditions of this investigation, the increase in total count attendant on the extrapolation of specific gravity from that of aggregate (S.G. = 1.5) to that of solid rock (S.G. = 2.7) is estimated, from the results of a related experiment^{*}, to be about 10%.

Accordingly, the results of measurements on the source aggregates may be extrapolated to those for solid rock filling the solid angle viewed by the detector by considering the pertinent thicknesses to be in the inverse ratio of the specific gravities of the respective materials. There will be an increase in intensity (up to 25%)^{**} attendant on the extrapolation. The 90% emission thicknesses so calculated for rock sources (S.G. = 2.7) are given in Table 2. In

^{*}As noted earlier, this density variation implies only a change in air distance, as the amount of absorber is constant. The intensity variation resulting from a change in air distance that would accompany an increase in specific gravity from 1.5 to 2.7 is estimated to average about 10% for thick sources. This estimate is based on the measured variation of intensity with air distance for a source of uranium aggregates (see footnote, page 20).

^{**}Sum of intensity variations due to density change (10%) and complete filling of solid angle of view (15%).

general, the surface layer comprising six to nine inches of rock (S.G. = 2.7) emits 90% of the gamma radiation of the infinitely thick rock source.

TABLE 2

Radioelement in Rock	Energy Band	90% Emission Thickness
(S.G. = 2.7)		
Potassium	"1.46 MeV" (1356 - 1566 keV)	6.5 inches
	740 - 1580 keV	minimum of 7.1 inches
Uranium	"1.76 MeV" (1700 - 1860 keV)	7.5 inches
,	210 - 2290 keV	minimum of 6.1 inches
Thorium	"2,62 MeV" (2480 - 2810 keV)	8.8 inches
	150 - 2810 keV	minimum of 7.8 inches

90% Emission Thicknesses for Rock Sources

For radiation from rock sources, porosity may be a significant variable affecting its intensity although not its energy distribution. However, a porous source saturated with a nonradioactive fluid (e.g. water) will have a lower intensity and a degraded spectrum compared with the unsaturated material. It is obvious, also, that allowance must be made for the density contrast, when using sources of crushed ore for calibration of radioactivity detectors that are used for quantitative measurements of "<u>in situ</u>" rock radioactivity.

- 29 -

GAMMA-RAY SPECTROGRAMS FROM ROCK SPECIMENS

Results of Experiments

The spectra of gamma radiation emitted from large rock specimens were measured in the laboratory with, to a first approximation, the same source-detector geometry as the prepared source aggregate, i.e., the rock samples filled the solid angle of view and, with allowance for density contrast, were each equivalent to one cubic foot of source aggregate.

Net rock spectrograms (i.e. with background subtracted) measured under these conditions are shown in Fig. 8 for the following rocks^{*}: St. George granite, Stanstead granite, Mount Royal nepheline syenite, and Sillery red slate. Rouyn gabbro was also measured, but the gross spectrum for a very long counting time was not significantly different from the laboratory background for an equivalent thickness of silica sand.

Photopeaks representing the major energies of the uranium and thorium series and of K-40 are recognizable in all spectrograms. Notable is the marked decrease in the count-rates in the high-energy part of the Stanstead granite spectrogram relative to the same part of the other rock spectrograms. This suggests a significant difference in the uranium and thorium concentrations of these rocks. Lesser spectral contrasts are also apparent^{**}.

* See Description of Sources, page 48.

** Note that two of the spectra are multiplied by constant factors for ease in presentation and comparison of all the spectra.





Comparison of these rock spectrograms with calculated spectrograms of unscattered gamma-rays from rock (5) shows the loss of resolution resulting from the scattering of photons in thick sources. Under such conditions of measurement, the energy lines most clearly distinguished against the Compton continuum are at 2.62 MeV in the thorium series, at 1.76 MeV in the uranium series and at 1.46 MeV for potassium-40. The 1.76 MeV line of uranium is not well resolved in the rock spectrograms; however, the more obvious 2.19 MeV line is difficult to use in analyzing a rock spectrogram, as this line is almost coincident with the thorium 2.20 MeV line and is also adjacent to a Compton edge in the thorium spectrum.

APPROXIMATE CONTRIBUTION OF EACH RADIOELEMENT TO THE GAMMA RADIATION FIELD FROM ROCKS

The separate contributions of uranium, thorium and potassium in the total field from rocks were estimated by comparing proportionately the spectrograms for the rocks and for the equivalent thicknesses of the standard source aggregates. The spectrograms for the thorium series and for the uranium series were "stripped off" the rock spectrogram as described subsequently. The specific conditions of measurement for aggregate and rock sources were to a first approximation equivalent. It is assumed that the uranium and thorium in the rocks are in secular equilibrium (as in the standards), for these rocks are early to middle Palaeozoic in age and were obtained from fresh quarry faces. As the potassium spectra were not investigated over the low-energy range (200-600 keV), except for the 6 inch thickness (Fig. 3), the required spectrogram for 12 inches of potassium standard was extrapolated in the low-energy range for this calculation. The extrapolation was based on some preliminary measurements with slightly different geometry but for the full energy range. Extrapolation was guided by the observed spectrogram for a thickness of 6 inches.

The calculation of the proportions of radiation from uranium, thorium and potassium in the total field emitted by St.' George granite is detailed in the following paragraphs. The rock source comprised several blocks with a basal area approximately 10 in. x 12 in. and an equivalent rock thickness of 6 inches.

Net integral count rates for given counting thresholds were calculated for each standard (12 inches of aggregate) and the St. George granite (6 inches of rock) by summing the count rates for individual channels within the given energy limits (see Table 3). Net count rates for selected energy lines were read from the spectrograms for these same sources (Table 4). These values were used in the following calculation:

Considering the 2.62 MeV energy line in the rock spectrogram (see Table 4), the count-rate due to thorium is $\frac{0.25}{6.9}$ times the count-rate of the thorium standard at the pertinent

TABLE 3

Comparative Integral Count-rates from Spectrograms of Standards and St. George Granite (Figures 3, 5, 6, 8)

	Standards (12 in. thick)			St. George Granite
Counting Threshold,	Th	U	K	
keV	c/m	c/m	c/m	c/m
200	2485	2147	368	365.0
500	1084	753	191	143, 2
1000	371	256	83	53.3
1600	192	59	0	11.6

TABLE 4

Comparative Count-rates for Selected Energy Lines in the Spectrograms of Standards and St. George Granite (Figures 3,5,6,8)

	Standar	ds (12 i	St. George Granite	
Energy Line,	Th	U	K	
MeV	c/m	c/m	c/m	c/m
2.62	6.9	0	0	0.25
1.76	5.3	6.4	0	0.59
1.46	6.3	7.4	4.6	2.74

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threshold (e.g., $\frac{0.25}{6.9} \times 2485 = 90.0 \text{ c/m at } 200 \text{ keV threshold}$).

For the 1.76 MeV energy line, the contribution due to Compton continuum from thorium is $\frac{5.3}{6.9} \times 0.25 = 0.19 \text{ c/m}$, and the contribution from uranium by difference is 0.59-0.19=0.40 c/m. Therefore, count-rate due to uranium is $\frac{0.40}{6.4}$ times count-rate of the uranium standard at the pertinent threshold

(e.g., $\frac{0.40}{6.4}$ x 2147 = 134.2 c/m at 200 keV threshold).

In the case of the 1.46 MeV energy line, the contribution due to Compton continuum from thorium is $\frac{6.3}{6.9} \ge 0.25 = 0.23$ c/m, that due to continuum from uranium is $\frac{7.4}{6.4} \ge 0.40 = 0.46$ c/m, and that due to potassium by difference is 2.74 - (0.23 + 0.46) = 2.05 c/m. Therefore, the count-rate due to potassium is $\frac{2.05}{4.6}$ times the countrate of the potassium standard at the pertinent threshold (e.g., $\frac{2.05}{4.6} \ge 368 = 164.0$ c/m at 200 keV threshold). The calculated rock activity is 90.0 + 134.2 + 164.0 = 388.2 c/m, whereas the measured activity was 365.0 (see Table 5).

The total count-rates obtained by summation of the calculated contributions of the individual radioelements agree fairly well with the values measured directly from the rock spectra. This suggests that the initial assumptions are valid, at least for the approximate calculations made here.

From Table 5 it is concluded that for the rocks analyzed

TABLE 5

Contribution of Radioelements to Rock Activity Measured with Various Counting Thresholds

Rock	Counting Threshold	Thor Contril	ium bution	Uraniu Contrib	ım ution-	Potassium Contribution		Total Rock Activity	
	keV	c/m	%	c/m	%	c/m	9%	Calculated	Measured
								c/m	c/m
St. George granite	200 500 1000 1600	90.0 39.2 13.4 7.0	23.2 22.8 20.2 65.4	134.2 47.1 16.0 3.7	$ \begin{array}{r} 34.6 \\ \overline{27.5} \\ \overline{24.1} \\ \overline{34.6} \end{array} $	164.1 85.5 37.1 0	$ \frac{42.2}{49.7} \\ \frac{55.7}{0} $	388.3 171.8 66.4 10.7	365.0 143.2 53.3 11.6
Stanstead granite	200 500 1000 1600	17.3 7.5 2.6 1.3	$ \begin{array}{r} 10.8 \\ \hline 9.7 \\ \hline 8.2 \\ \hline 61.9 \\ \end{array} $	28.8 10.1 3.4 0.8	$ \begin{array}{r} 17.9 \\ \hline 13.1 \\ \hline 10.7 \\ \overline{38.1} \\ \end{array} $	114.4 59.4 25.8 0	$\frac{\begin{array}{c} 71.3\\ \overline{77.2}\\ \overline{81.1}\\ \underline{0} \end{array}$	160.5 77.0 31.8 2.1	142.0 70.6 30.4 2.1
Mount Royal nepheline syenite	200 500 1000 1600	96.9 42.3 14.5 7.5	26.1 25.1 21.9 73.5	96.6 33.9 11.5 2.7	$ \begin{array}{r} 26.0 \\ \hline \hline \hline \hline \hline \hline \hline \hline \hline \hline $	177.7 92.3 40.1 0	$\frac{47.9}{54.8}\\ \frac{60.7}{0}$	371.2 168.5 66.1 10.2	324.0 165.0 51.4 10.1
Sillery slate	200 500 1000 1600	50.4 22.0 7.5 3.9	22.4 21.3 18.3 70.9	57.1 20.0 6.8 1.6	$ \begin{array}{r} 25.3 \\ \hline 19.4 \\ \hline 16.6 \\ \hline 29.1 \\ \end{array} $	117.7 61.1 26.6 0	$ \frac{52.3}{59.3} \\ \frac{59.3}{65.1} \\ \underline{0} $	225.2 103.1 40.9 5.5	213.8 95.4 41.4 5.7

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and for counting thresholds between 200 keV and the primary photopeak energy of K-40 (1.46 MeV), the radiation from potassium predominates over that from uranium and from thorium. Potassium contributes 40 to 80% of the total count depending on the rock type and the counting threshold chosen. Of the balance, the contribution from uranium is slightly greater than that from thorium. As counting thresholds are raised above the primary energy of potassium, the contribution from thorium becomes increasingly more dominant over that from uranium. For such high threshold energies there is, of course, no contribution from potassium.

The predominance of radiation from potassium in the field of average rocks was postulated previously by Gregory (13, Table II) from calculations based on measurements by Russell (14). However, the uranium contribution so calculated is much smaller than that measured for the rocks in Table 5. This discrepancy in part results from differences in the concentration of radioelements between the average rocks in the previous calculations and the rocks that were measured in the work reported here. However, the main difference between the calculated and the measured uranium contribution to the field of average rocks is attributed to the use in the calculations of Russell's value for the activity of uranium. This value now appears to be low relative to his values for the activity of thorium and potassium (see Table 6).

The measured activity of a source varies with the technique

of measurement because it is a function of the source-detector geometry and the detector characteristics. However, the results of different investigations may be compared with each other if the conditions of measurement are similar. Activity ratios $(A_{K}:A_{U}:A_{Th})$ calculated for unit amounts of the calibration standards of other investigations similar to the present work are compared by normalizing the ratios to $A_{K} = 1$ (Table 6). As the techniques of measurement are not all exactly the same, small variations in the ratios may be expected.

TABLE 6

Investigator	A _K :A _U : A _{Th}	Apparatus
Russell, W.L.(14)	1 :2800:2300	Geiger counter, thin annular source surrounding detecting element.
Slack, H.A.(15)	1 :4950:2470	Multiple Geiger counters.
McCallum,G.J.(16)	1 :4300:2200	Geiger counter, conditions of measurement not stated.
This report	1 :4600:2000	*Scintillation counter with 200 keV counting threshold; source 12 inches thick and 6 inches from detector.

Activity Ratios for Unit Amounts of Radioelements (normalized to A_K = 1)

* The ratios A_K:A_U:A_{Th} vary with the counting threshold and the source-detector geometry. For the present work the ratios at 500 keV threshold are 1:3400:1700 and at 1000 keV, 1:2400:1300. For a much smaller sample placed close to a similar detector, the activity ratios calculated from measurements by Horwood (17, Table 1) are 1:8615:4559 at 200 keV threshold; 1:4802:2571 at 500 keV; and 1:3100:1516 at 1100 keV.

The value of A_U calculated from Russell's results is low (about 60% of the other A_U values), while his value of A_{Th} is similar to the other A_{Th} values. This discrepancy probably results from disequilibrium in the carnotite that was used as the uranium standard (18). It is known from theory and assaying practice, that in general the specific activity of uranium in equilibrium is about twice that of thorium in equilibrium for low counting thresholds. This relationship is also indicated by the results in Table 6 excepting those calculated from Russell's calibrations. These facts support the previous conclusion that the calculated contribution from uranium to the gamma radiation field of average rocks is too small because Russell's value for the activity of uranium is low.

The rocks used in this experiment have radioelement concentrations (see Table 7) that approximate those of common rocks (19, 20), excepting carbonates. However, the Th/U ratios for these rocks are lower than average (19). From these measurements and the previous calculations it is concluded that, for common rocks, the intensity of gamma radiation from potassium predominates over that from either uranium or thorium and may exceed their sum. The relative proportions of the radiation field due to uranium and thorium will depend on the Th/U ratio of the given rock. For a Th/U ratio of about 2, uranium and thorium will have equal significance as sources of gamma radiation. Average rocks have a Th/U ratio of 3 to 4 and accordingly the emitted radiation from thorium should generally exceed that of uranium by a factor of about two. In addition to the dependency on radioelement concentration, the measured contributions will also depend on the characteristics of the detector.

In contrast to common rocks, soils may show great variability in the relative contributions to the radiation field due to uranium, thorium and potassium, because of leaching and deposition of these radioelements or a change in their state of equilibrium during or after soil formation^{*}.

THE POSSIBILITY OF "IN SITU" DETERMINATION OF THE RADIOELEMENT CONTENT OF ROCKS

The calculation of the contribution of each radioelement to the gamma radiation field (page 33) may logically be carried one step further to an actual evaluation of the radioelement content of the rocks if the contents of the standards are accurately known. This is done in an analogous calculation by substituting the known radioelement content of the standard for the measured activity of the standard. Thus, for the St. George granite (Table 5) the thorium content is $\frac{0.25}{6.9} \times 350 \times 10^{-6}$ g/g = 12.7 x 10⁻⁶ g/g^{**}. Because of the geometrical approximations previously discussed, the calculated content of the radioelements in the rock can only be approximate.

Telfair et al. (21) estimate that about 20% of the gamma activity of the Miami silt loam is contributed by potassium. The balance of the radiation is attributed to uranium and thorium.

The thorium standard contains 0.040% ThO₂ = 350 x 10^{-6} g Th/g.

- 40 -

The results calculated from the rock source spectra agree fairly well with the analyses determined by other methods (see Table 7). This agreement suggests that, with an improved technique, it may be possible to perform analytical determinations of uranium, thorium and potassium on rock specimens, in boreholes, or on suitable rock outcrops. Such a technique would require calibration standards of similar density to the rock and a greater constancy of source-detector geometry than was achieved in this analysis. Measurements on a rock surface or in a bore-hole would have the advantage of exposing the detector to a much larger sample of rock than would a laboratory determination. Sourcedetector geometry would be constant with effective infinity of both thickness and surface extent of the source. A further improvement could be expected from reducing the background count-rate of the measurement. Finally, energy lines other than those used herein should be investigated to see whether they will improve the accuracy of the determination.

- 41 -

In this study, background was relatively high because the sources were placed over the open top of the lead castle. Thus, background included a significant amount of radiation from radioelements in the surrounding building materials as well as cosmic, atmospheric and instrument activity.

Rock	Th, 10-6 g/g		U,10-	-6 g/g	K,10 ⁻² g/g		
ROCK	A ₁	A 2	A ₁	A ₂	Al	A ₂	A ₃
St. George granite	12.7	25.	8.1	10.	4.6	4.4	puti
Stanstead granite	2.4	-	1.7	-	3.2		-
Mount Royal nepheline syenite	13.6	20.	5.9	11.	5.0	3.7	4.7(a)
Sillery slate	7.1		3.5	-	3,3	-	2.8(b)

Comparative Radioelement Analyses of Four Rocks

A₁ - analysis from thick source spectra, this report.

A₂ - gamma-ray spectroscopic analysis of pulverized rock by J.L. Horwood.

A₃ - chemical analysis from the literature as noted.

- (a) J.A. Dresser and T.C. Denis. Geology of Quebec, Vol. II, Descriptive Geology; Quebec Department of Mines, Report No. 20, p. 464 (1944).
- (b) J.A. Dresser and T.C. Denis, op.cit., p. 390.

CONCLUSIONS

- Rocks and soils that contain uranium, thorium and potassium emit characteristic gamma-ray spectra that reflect the proportions of these radioelements present in significant quantities in the source. In detail, the spectrograms observed are specific for the particular detector and conditions of measurement, but useful generalizations may be made.
- 2. The major photopeaks of all the radioelements present in a source are recognizable in gamma-ray spectrograms measured for sources ranging in thickness from one-half inch to effective infinity.
- 3. The Compton continuum of the spectrogram of a thick source includes the significant effects of secondary photons from Compton scattering in the thick source, as well as those effects generally observed from such Compton interactions within the detector.
- 4. With an increase in source thickness, the yield of source-scattered photons increases in importance, but does not overwhelm the major photopeaks at any thickness. The radiation approximates a constant energy distribution at great thicknesses.

- 5. The thickness of source material that effectively contributes to the spectrum emitted from the surface of a thick source varies with the porosity of that source. Thus, 12 to 18 inches of aggregate (S.G. = 1.5) or 6 to 9 inches of rock (S.G. = 2.7) emit 90% of the intensity of an infinitely thick source of the same material.
- 6. In the gamma radiation field of average rocks, radiation from potassium predominates over that from either uranium or thorium. The proportions of the field that are emitted by uranium or thorium vary with the Th/U ratio of the rock. In general, radiation from thorium will exceed that from uranium. The actual contribution of each radioelement to the measured radiation intensity depends on the counting threshold of the detector(discriminator level). If the threshold is set to accept gamma-rays below 1.46 MeV, so that primary potassium radiation is counted, then 40% or more of the measured intensity is due to radiation from potassium and the balance comes from uranium and thorium in proportions dependent upon the Th/U ratio.
- 7. With suitable calibration sources and standardized source-detector geometry, the technique of gamma-ray spectroscopy may have useful quantitative aspects

- 44 -

for the non-destructive analysis of rock specimens for the elements U, Th and K. Another possible and potentially useful analytical technique might be developed with instrumentation adapted for measurements on the face of a rock outcrop or in a bore-hole.

- Gamma-ray spectrum analysis should prove to be a valuable technique in geological correlation.
- 9. Further investigations should use larger sources (perhaps rock outcrops) in order to preclude the geometrical approximations encountered in this investigation.

ACKNOWLEDGMENTS

This investigation was conducted in the Physics and Radiotracer Laboratories of the Mineral Sciences Division, Mines Branch, Department of Mines and Technical Surveys in Ottawa. The authors are indebted to A.L. Fournier and R.M. Hopwood, who made many of the measurements and assisted in the computations. Analysts and suppliers of source materials are acknowledged in Appendix A.

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APPENDIX A

DESCRIPTION OF SOURCES

The aggregate sources were placed in wooden boxes, of which the tops and bases were fir plywood ($\frac{1}{4}$ inch thick) and the sides were pine boards (3/4 inch thick). The boxes were designed to give a constant basal source area (12 inches square) and source thicknesses of $\frac{1}{2}$, 1, 2, 4 and 6 inches. Most boxes were of the larger size.

Potassium sources: Feldspar aggregate $(-\frac{1}{2} \text{ inch mesh})$ averaging 12.4% K₂O (Analyst: E.M. Penner, Mines Branch). No thorium or uranium was detected by gamma spectroscopy of a pulverized sample. The bulk specific gravity was 1.6. The feldspar was obtained from the Back quarry, Buckingham, Quebec, and was donated by the International Minerals and Chemical Corporation (Canada) Ltd., courtesy of W.H. Hood, general manager, and L. Joyce, plant superintendent.

Uranium sources: Finely crushed pitchblende ore was thoroughly mixed in a dominant matrix of glass sand. The average uranium content of the mixture was $0.015\% U_3O_8$ (beta-gamma radiometric analysis by C. McMahon, Mines Branch). The bulk specific gravity was 1.5. Thorium and potassium, which were present in very small amounts in the pitchblende ore, were not detectable in the gamma spectrum of the source aggregate. The # 35 mesh glass sand (SiO₂:99.7%+) was supplied by Dominion Silica Corporation Ltd., Lachine, Quebec, from quarries at St. Donat, Quebec. No uranium, thorium or potassium was detected by gamma spectroscopy of the sand.

In addition, bags of pitchblende concentrate $(-\frac{1}{2} \text{ inch mesh})$ averaging 15.5% U₃O₈, 0.003% ThO₂ and 0.06% K (J.C. Ingles, Head of Control Analysis Section, Mines Branch) were used in a related series of experiments, in which crude thick-source measurements were made with this material located at a source distance of 22 feet in air. The pitchblende concentrate (bulk specific gravity = 1.8) originated from Port Radium, N.W.T., and was loaned by Eldorado Mining and Refining Ltd., Ottawa, Ontario, through the courtesy of W.M. Gilchrist, president.

Thorium sources: Finely crushed thorite ore was thoroughly mixed in a dominant matrix of glass sand. The average thorium content of the mixture was 0.040% ThO₂ (beta-gamma radiometric analysis, C. McMahon, Mines Branch). The bulk specific gravity was 1.4. Uranium and potassium, which were present in very small amounts in the thorite ore, were not detectable in the gamma spectrum of the source aggregate.

In addition, bags of thorite concentrate $(-\frac{1}{2} \text{ inch mesh})$ averaging 14.3% ThO₂, 0.13% U₃O₈ and 0.31% K (J.C. Ingles, Head

-49 -

of Control Analysis Section, Mines Branch) were used in a related series of experiments, in which crude thick-source measurements were made with this material at a source distance of 22 feet in air. The thorite ore (bulk specific gravity = 1.3) originated from an occurrence near the boundary between Broadwater and Meagher counties in Montana and was supplied by Davison Chemical Company, Pompton Plains, N.J.

Rock sources: Large single blocks or several closefitting pieces of rock, averaging about 7 inches in thickness, were used. The rock specimens, collected under the direction of C.H. Gauthier of the Geological Survey of Canada, comprised: 1. Pink hornblende granite, uear St. George, N.B.; 10 in. x 12 in., comprising several pieces with an equivalent rock thickness of 6 inches.

2. Grey biotite granite, Graniteville quarries, Stanstead county,

3. Grey nepheline syenite, Mount Royal, Montreal, Quebec; rough block, approximately 9 in. x 8 in. by 7 in. thick.

Quebec; squared block, 7 in. x 7 in. by $7\frac{1}{2}$ in. thick.

4. Red slate, Sillery formation, Actonvale, Quebec; group of flat, tabular slabs approximately 10 in. x 12 in. in size with an equivalent rock thickness of 6 3/4 inches.

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5. Greenish-black gabbro, Rouyn, Quebec; an irregular block with flat face, approximately 8 in. x 9 in. by 6 in. thick.

APPENDIX B

INSTRUMENTATION

The gamma-ray detector consisted of a crystal of thallium-activated sodium iodide (2 inches thick by 2 inches in diameter) mounted directly against the window of a photomultiplier tube (R.C.A. Type 6655). The crystal was shielded from light and moisture by a magnesium cylinder cemented over the phototube. A thin layer of alumina was packed around the crystal to serve as a light reflector and aluminum foil was cemented to the end of the cylinder to serve as a window between source and crystal. Four inches of lead shielding were placed around the detector to minimize background radiation. The castle aperture was 4 inches square and was located 5 3/4 inches above the top face of the crystal (Fig. 1). The resolution of the detector, measured for the 0.662 MeV photopeak of Cs-137, was 10%.

The pulses from the output of the phototube were fed through a cathode follower in the detector assembly, then amplified 60 times, and sorted according to amplitude in a Goulding, 100-channel, pulse-height analyzer (Computing Devices of Canada Ltd., Ottawa, Ont., Model AEP-2230). The high voltage supplied to the phototube was stabilized by a string of fifteen Mullard OG 3 glow-tubes, tapped between tubes (i.e. at 85-volt intervals) for coarse adjustment, with an additional vernier voltage adjustment by a ten-turn Helipot potentiometer controlling a 200-volt electronically-regulated power supply connected in series between the glow-tubes and ground. This latter control was useful for making minor corrections in phototube gain when necessary.

The amplitude spectra were displayed on the cathoderay tube of the analyser and were also plotted graphically on a Varian recorder (Model G-10). The spectra were related to an energy scale by using standard sources of Co-57, Na-22, Cs-137, Mn-54 and Co-60. Major peaks in the spectra of the aggregate sources (potassium, natural thorium and natural uranium) served as internal standards, whereby any changes in gain resulting in a shift in the energy scale could be observed and corrected. Change in gain with change in count-rate was not observed in this work, because low-activity sources and standards were used and the phototube was selected to minimize this effect.

Good overall stability in the equipment was obtained by using a Sorensen regulator to stabilize the A.C. input to the high voltage supply, the pulse amplifier, and recorder. Temperature and humidity were maintained relatively constant in the laboratory.

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