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THE DEVELOPMENT AND APPLICATION OF A BETA-GAMMA-GAMMA
MULTIDIMENSIONAL SPECTROMETER⁵

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SUMMARY

A beta-gamma-gamma multidimensional spectrometer has been developed which employs two 12-inch diameter by 8-inch thick NaI(Tl) crystals with associated 4-inch thick NaI light pipes and two 5-inch diameter by 1/2-inch thick gas proportional counters as principal detectors and which utilizes two 40-inch diameter by 24-inch thick plastic phosphors as anticoincidence shields. The detector system is coupled to two 4096 word computer memories which record gamma-ray events in coincidence with either beta detector. The gamma-ray energies identify the radionuclide while the beta detectors can identify the relative position of the radioactivity in the specimen being analyzed. The background and efficiency of the system and its application to the identification of radionuclides in environmental, biological, and extraterrestrial materials are discussed.

Introduction

A variety of methods are available to the nuclear scientist for the analysis of radionuclides, and some of these are truly radionuclide specific and nondestructive. The nondestructive methods commonly applied to analytical schemes utilize gamma-ray spectroscopy in a variety of coincidence and anti-coincidence modes. Some of these techniques, which employ primarily Ge(Li) and NaI(Tl) detector systems, are used to investigate the low level radionuclide concentrations in atmospheric, oceanographic, terrestrial, and extraterrestrial specimens. In some cases it is desirable to know the fraction of radionuclides on the surface of a sample. Standard methods do not provide radionuclide depth distributions, and only through use of a technique involving the measurement of beta and/or alpha emissions can information on the depth of a radionuclide be determined.

In recent years many significant advances have been made in the development of low-level radionuclide analysis systems. For low-level gamma-ray spectrometry, large-volume NaI(Tl) crystal multidimensional gamma-ray spectrometer systems have been developed and applied in the nondestructive measurements of complex spectra of radioisotopes distributed throughout large samples. These sensitive and versatile systems have been used extensively for precise radionuclide measurements in terrestrial, oceanographic, atmospheric and extraterrestrial material. A modification of this type of system requiring beta-coincidence has provided a selective and sensitive means for radionuclide analyses of weightless samples and of the surface of materials such as environmental and biological specimens, neutron irradiated bone, and Apollo and Surveyor lunar specimens. This newly developed system allows the identification of radionuclides through their characteristic gamma-ray emissions and provides

information on their surface and depth distribution through their associated charged particle emissions. The system can also measure radionuclides throughout the entire specimen in the normal manner.

Instrumentation

The beta-gamma-gamma multidimensional spectrometer system (Fig. 1) consists of two 12-inch diameter by 8-inch thick NaI(Tl) crystals with associated 12-inch diameter by 4-inch thick pure NaI light pipes and two 5-inch diameter by 1/2-inch thick methane gas operated proportional beta detectors with 500 microgram per cm² gold plated mylar windows. The large crystals were chosen to maximize efficiency for large environmental samples while providing space for the large-area beta detectors. The light pipes serve to shield the active crystal from background originating in the phototubes or exterior cave surfaces. Each crystal has seven 3-inch RCA 8054 phototubes with low background tube bases. The upper and lower crystal resolutions at the 0.662 MeV ¹³⁷Cs line are 9.6% and 10.3% fwhm, respectively. The beta detector thresholds start at 3000 volts with slopes of <1.5%/100 volts, and the detectors are a maximum of 47% efficient for a massless source of ⁹⁰Sr.

The spectrometer system is electronically shielded by two 40-inch diameter by 24-inch thick plastic phosphors and by one 16-inch diameter by 12-inch thick plastic "end plug" operating in anticoincidence. The entire assembly is mass shielded from external radiation with four inches of lead and four inches of paraffin containing five weight percent boron. A specimen to be analyzed is placed between the two beta detectors which themselves are inserted between the two primary gamma-ray detectors. As the lower beta detector recognizes an electron the two NaI(Tl) crystals are allowed to record single and coincidence gamma-ray events in a 4096 channel memory operating in a 64 by 64 channel array. Coincidence gamma-ray events are stored in the energy-energy plane of the memory according to their photon energies while single gamma-rays which interact in either crystal are stored in a normal manner on the axis of the array. A similar recording of gamma-ray events occurs in a second 4096 channel memory when a beta particle is detected in the upper beta system. When events are recorded simultaneously by a primary gamma ray detector and the anticoincidence shield, a blocking pulse is applied to the entire system and no gamma-ray events are recorded (Fig. 2). Since beta particles have a short range in solid materials, those which are detected must originate from radionuclides near the surface of the sample. Thus, in the beta-gamma-gamma mode of operation, the radionuclides are identified by their coincident gamma-rays and are measured from shallow depths within the specimen. By turning off the beta gate requirements, the system is responsive to gamma-ray emitting radionuclides from the whole sample.

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Efficiency, Background, and Sensitivity

The efficiency of the present system for a point source of gamma radioactivity is shown in Fig. 3, both with and without the beta counters in position. Also shown for comparison is the efficiency of 3-inch diameter by 3-inch thick crystals in a similar arrangement. The counting efficiency, $2E$, is defined as the fraction of the gamma-rays emitted by a source that are totally absorbed in either detector. The total coincidence efficiency, $2E_1E_2$, is the probability of total absorption of two gamma rays, one in each of the two detectors. The beta detectors are each 47% efficient for ^{90}Sr ($\beta_{\text{max}} = 0.546$ MeV) and even 33% efficient for ^{210}Po ($\alpha = 5.305$ MeV).

Low-level radionuclide measurement systems require both low backgrounds and high counting efficiencies. As shown in Table 1, the backgrounds are reduced very substantially with a minimal loss of efficiency for a weightless sample by requiring a beta coincidence signal. The net result is an increase in sensitivity typically about two orders of magnitude for coincidence gamma-rays and even more for low energy single gamma-ray events. Table 2 lists the minimum detectable activities as a function of gamma-ray energy based on the following equations from Curie⁽¹⁾:

$$A_d = \frac{2.71 + 4.65 \sqrt{\text{Bkgd}}}{(\text{eff}) (\Delta T)}$$

$$A_Q = \frac{50 \left\{ 1 + \sqrt{1 + \frac{\text{Bkgd}}{12.5}} \right\}}{(\text{eff}) (\Delta t)}$$

where

A_d = the minimum detectable activity (95% confidence level),

A_Q = the activity required to obtain a precision of 10%,

and

Bkgd = total background in a counting period, ΔT , in a system with efficiency, eff.

The assumptions are: that the radioactive specie has a long half-life such that $1 - e^{-\lambda \Delta t} \rightarrow \lambda \Delta t$, that the sample is thin (<0.1 inches) and no more than 5-inches in diameter, and that the counting interval is 10,000 minutes. These calculations show that a beta coincidence requirement allows a smaller quantity of a single gamma-ray emitting isotope to be detected than any isotope possible through ordinary gamma-gamma coincidence techniques. They also demonstrate that there is very little dependence on gamma-ray energy or even on coincident gamma-rays as long as there is a beta gate requirement imposed. In effect, the background has been reduced so low (less than 1 count in 30,000 minutes for some isotopes) that the limiting feature of the analysis is simply obtaining a significant number of decays in a reasonable time period to allow a quantitative statement to be made about the activity levels.

Ultra Low Level Counting Applications

The determination of low concentrations of radionuclides in mixtures of environmental, biological, and extraterrestrial materials requires extremely sensitive analytical procedures and counting equipment. To provide the required sensitivity, gamma-ray spectrometers employing NaI(Tl) detectors were normally used in conjunction with radiochemical separation procedures. For most ultra low level measurements Ge(Li) diodes do not yet provide the required sensitivity. With the advent of multidimensional NaI(Tl) gamma-ray spectrometers²⁻⁴ the simultaneous determination of up to 20 radionuclides in air, water, and biological specimens became possible. The use of β - γ - γ coincidence techniques, as described in this paper, significantly improve the sensitivity and selectivity of environmental material analyses. The present β - γ - γ spectrometer allows trace radionuclide concentrations to be determined in a variety of trophic levels and thus to be followed along various biological and physical transport pathways from the release of the radionuclides to their ultimate fate.

Fallout radionuclides include a complex mixture of old nuclear weapon debris interspersed with new short-lived debris from more recent nuclear tests and cosmic-ray interactions in the atmosphere. This situation frequently produces complex mixtures of radioisotopes which could not be measured by available instrumental methods of analysis. The present β - γ - γ spectrometer system allows many of the trace constituents of cosmogenic, fallout, and reactor effluent radionuclide mixtures to be detected in the presence of 100-fold or greater concentrations of interfering gamma ray emitting radionuclides. For example, ^{106}Rh the daughter of long-lived ^{106}Ru , is normally analyzed through its coincident 0.51-0.62 MeV gamma-ray cascade. The large NaI(Tl) detectors used to measure low-level activities cannot adequately resolve this peak from that of positron annihilation, 0.51-0.51 MeV, and in many cases the determination of ^{106}Rh has been voided by relatively small quantities of annihilation or Compton scattered events. The β - γ - γ spectrometer which provides separate $\beta\gamma\gamma$, $\beta\gamma$, $\gamma\gamma$ and γ spectra effectively removes many interfering gamma rays from the spectrum and permits a much more sensitive analysis of ^{106}Rh to be completed.

High concentrations of ^{65}Zn often interfere with the analysis of fallout or reactor produced $^{95}\text{ZrNb}$ which is normally determined by measuring the 0.724, 0.756, and 0.765 MeV single emission gamma rays. Zinc-65, which has a gamma ray at 1.115 MeV, can yield sufficient Compton-scattered events to obscure the peak from $^{95}\text{ZrNb}$. Since ^{65}Zn does not decay by beta emission to the 1.115 MeV level, it is not stored in the $\beta\gamma\gamma$ spectrum and its interference is successfully eliminated.

Other radioisotopes frequently masked in reactor effluent water or air filters containing fallout debris include $^{140}\text{BaLa}$ and ^{22}Na which can be determined at much lower levels in the β - γ - γ spectrometer. Sodium-22 is produced in the atmosphere by cosmic ray spalla-

tion with argon nuclei, and precise measurements of this isotope are important for studies such as those determining the atmospheric residence time of natural aerosols. Small quantities of radioisotopes such as ^{239}Np , ^{155}Eu , ^{144}Ce , ^{131}I , ^{122}Sb , ^{103}Ru , ^{99}Mo , ^{76}As , and ^{65}Ni , which decay by a beta gamma coincidence mode, can also be directly measured in fallout or reactor effluents in the presence of large quantities of noncoincident beta emitting or electron capture radioisotopes such as ^{137}Cs , ^{65}Zn , ^{54}Mn , ^{51}Cr , and ^7Be with this new spectrometer system.

In instances where the daughters of thorium or uranium are being used as tracers of air masses, the $\beta\gamma\gamma$ spectrometer facilitates their direct and simultaneous measurement. For example, ^{212}Pb , ^{212}Bi , and ^{208}Tl are all members of the ^{232}Th decay chain. These isotopes have half-lives of 10.64 hours, 60.6 minutes, and 3.1 minutes respectively, and a simultaneous measurement of their ratios permits a determination of the elapsed time since they were last in equilibrium. This technique is providing a method to study precipitation scavenging of air masses and the deposition patterns of particles in uranium mine atmospheres.

The same system can also be utilized to measure elemental surface concentrations by the analysis of the gamma-ray emitting radionuclides produced by neutron or proton irradiations. In Fig. 4 various spectra of the ^{60}Co in a bone specimen are shown. The $\gamma\gamma$ coincidence spectrum of ^{60}Co provides the concentration of the whole bone specimen. The $\beta\gamma\gamma$ system spectrum provides the surface concentration of ^{60}Co . Absorbers can be placed between the sample being analyzed and the beta detector to determine the depth gradient of the radioactive element of interest.

In Table 3 the depths at which concentration gradients of many pollutants can be determined for biological material such as bone are given. The first element, Hg, which may plate on the bone surface causing osteomyelitis, can be determined at very shallow depths. For example, the beta particle from ^{203}Hg , a neutron activation product, has a predominant maximum beta energy of 0.214 MeV. Since its range in tissue-like materials is about 40 milligrams per square centimeter, it can only be detected through a depth of about 0.25 millimeters. The range of the beta particle defines the depth from which gamma-ray events are recorded. In this application interference from electron capture radionuclides is eliminated since these events are stored in the single or coincidence gamma-ray spectra. With this technique the deposition pattern within a specimen of toxic trace materials can be determined. Utilization of this instrument is also being employed to determine deposition patterns of radionuclides on selected surfaces of environmental materials relative to their concentrations in the whole specimen.

In addition to studies of terrestrial materials, the system is being used to measure ^{22}Na and ^{26}Al on the surface of returned lunar specimens. At or near the lunar sur-

face the proton flux is intense and produces concentrations of radionuclides from (p,xpyn) reactions on Mg, Al and Si significantly larger than are produced a few millimeters deep by the lower flux of protons with energies exceeding about 50 MeV. This depth variation can be measured by the beta-gamma-gamma coincidence method. Such measurements document the most recent orientation of the specimen on the lunar surface, any recent changes in this orientation and its degree of surface erosion. In Table 4 concentrations of ^{26}Al from various Apollo 12 lunar specimens analyzed in the beta-gamma-gamma coincidence system are shown. The data are based on the positron counting rates which were triggered by beta particles from either the upper or lower surface of the specimen.

In Fig. 5, the spectrum of a specimen with a homogenous activity is compared schematically to that obtained from a specimen with a radioactivity gradient. In the case of a partially buried rock which would therefore have no radioactivity gradient on its bottom side, the lower beta detector would not indicate a high surface concentration. The upper beta gated spectra would show high concentrations of ^{26}Al on the surface of the rock. Specimens 12051,1 and 12016,0 show comparable ^{26}Al on their upper and lower surfaces, indicating that the average surface presented to each beta detector must have had an equal proton bombardment. However, in the cases of rock fragments 12051,3 and 12005,0 a significant variation is observed indicating that the rock face with the higher activity was oriented upward on the lunar surface.

References

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5. This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1).

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TABLE 1

Background Counting Rates and Sensitivities for
 ^{22}Na and ^{26}Al Gamma-Rays in the Multidimensional
 Spectrometer System With and Without Beta Gate Requirements

Energy (MeV)	Beta-Gamma-Gamma		Gamma-Gamma	
	Background (cpm)	Sensitivity* (cpm ⁻¹)	Background (cpm)	Sensitivity* (cpm ⁻¹)
0.511	3.34×10^{-2}	6.08×10^{-3}	108	2.41×10^{-6}
1.275	1.85×10^{-2}	1.72×10^{-1}	43.2	1.44×10^{-4}
1.81	1.32×10^{-2}	3.47×10^{-2}	15.9	4.37×10^{-4}
0.511 by 0.511	3.8×10^{-3}	4.2×10^{-1}	0.182	1.16×10^{-2}
0.511 by 1.275	1.8×10^{-3}	9.1×10^{-1}	0.102	1.54×10^{-2}
0.511 by 1.81	5×10^{-4}	1	0.098	9.5×10^{-3}
0.511 by 0.511 by 1.275	5×10^{-4}	30	0.098	1.4×10^{-1}
0.511 by 0.511 by 1.81	3×10^{-4}	10	0.033	2.4×10^{-1}

* Defined as the Square of the Point Source Efficiency as a
 Decimal Divided by the Background, [(2E)²/B].

TABLE 2

Minimum Detectable Activities in γ/Min .

Energy (MeV)	Beta-Gamma-Gamma		Gamma-Gamma	
	A_d	A_Q	A_d	A_Q
0.5	0.0206	0.0720	1.09	3.32
1.0	0.0125	0.0502	0.750	2.29
1.5	0.0122	0.0511	0.713	2.18
2.0	0.0128	0.0541	0.547	1.67
2.5	0.011	0.049	0.555	1.70
0.5 by 0.5	0.026	0.12	0.175	0.565
1.0 by 1.0	0.009	0.1	0.073	0.27
1.5 by 1.5	0.02	0.2	0.04	0.2
2.0 by 2.0	0.09	0.2	0.07	0.3
2.5 by 2.5	<0.01	0.2	0.03	0.2

TABLE 3

Trace Elements and the Relative Depths
at Which They May Be Determined in Bone

Element	Radionuclide	Predominant Beta Emission Max. Energy (MeV)	Average* Endpoint Energy (MeV)	Range In Al (mg/cm ²)	Depth mm
Hg	²⁰³ Hg	0.214	0.214	46	0.25
Co	⁶⁰ Co	0.319	0.319	85	0.46
Zn	⁶⁵ Zn	0.325	0.325	87	0.47
Sc	⁴⁶ Sc	0.357	0.357	95	0.51
Fe	⁵⁹ Fe	0.475 (51%) 0.273 (48%)	0.378	113	0.61
Br	⁸² Br	0.444	0.444	140	0.76
Cs	¹³⁴ Cs	0.662 (71%)	0.499	165	0.89
Ca	⁴⁷ Ca	0.67 (82%)	0.67	255	1.38
Rb	⁸⁶ Rb	0.71	0.710	265	1.43
Au	¹⁹⁸ Au	0.962	0.962	380	2.05
Sb	¹²⁴ Sb	0.61 (49%) 2.317 (21%)	1.019	420	2.27
Na	²⁴ Na	1.389	1.389	500	2.70
K	⁴² K	1.97	1.97	925	5.00
As	⁷⁶ As	2.41 (31%) 1.76 (16%)	2.075	950	5.13
Cr	⁵¹ Cr	E.C.			
Se	⁷⁵ Se	E.C.			

* Weighted average of the maximum beta energies for those leading to excited gamma-ray decay states or annihilation radiation.

TABLE 4

Surface Concentration of ^{26}Al in Apollo 12 Specimens

Rock	^{26}Al dpm/kg*		^{26}Al dpm/kg**		Upper/Lower
	Upper Beta Gate	Lower Beta Gate	Upper Beta Gate	Lower Beta Gate	
12051,1	160 $\pm$ 20	159 $\pm$ 18	130 $\pm$ 17	127 $\pm$ 15	1.0 $^{+.3}_{-.2}$
12051,3	242 $\pm$ 18	120 $\pm$ 10	193 $\pm$ 15	95 $\pm$ 8	2.0 $^{+.4}_{-.3}$
12005,0	164 $\pm$ 17	91 $\pm$ 12	137 $\pm$ 14	77 $\pm$ 11	1.8 $^{+.5}_{-.4}$
12016,0	135 $\pm$ 16	119 $\pm$ 12	109 $\pm$ 13	93 $\pm$ 10	1.2 $^{+.3}_{-.3}$

* Assuming all the β^+ is ^{26}Al with a negligible contribution from ^{22}Na .

** Assuming the β^+ fraction has an $^{26}\text{Al}:^{22}\text{Na}$ ratio of 2.5:1.

CAPTIONS

- Fig. 1 A beta-gamma-gamma multidimensional spectrometer system
- Fig. 2 Electronics block diagram of a beta-gamma-gamma multidimensional spectrometer system
- Fig. 3 Singles and coincidence counting efficiencies of beta-gamma-gamma and gamma-gamma multidimensional spectrometer systems
- Fig. 4 Determination of the surface and total concentrations of beta emitting neutron activated radionuclides in bone by nondestructive beta-gamma-gamma multidimensional spectrometry
- Fig. 5 A beta-gamma-gamma spectrum of lunar rock with and without a radioactivity gradient

BETA DETECTOR
COINCIDENCE OFF;
X DPM ^{60}Co UNIFORMLY
DISTRIBUTED THROUGHOUT
THE SAMPLE

BETA DETECTOR
COINCIDENCE ON;
X DPM ^{60}Co UNIFORMLY
DISTRIBUTED THROUGHOUT
THE SAMPLE

BETA DETECTOR
COINCIDENCE ON;
X DPM ^{60}Co LOCATED
ON THE SURFACE
OF THE SAMPLE

UPPER BETA DETECTOR
GATED SPECTRUM

LUNAR ROCK SHIELDED
FROM SOLAR RADIATION
(FAIRLY HOMOGENEOUS
RADIOACTIVITY)

SURFACE
CONCENTRATION
OF ^{26}Al

LUNAR ROCK EXPOSED
TO SOLAR RADIATION
(INCREASED SURFACE
RADIOACTIVITY)

LOWER BETA DETECTOR
GATED SPECTRUM

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