MAY 14 1965

BNWL-SA-119
BNSA-119

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

CONF-650507-35

MASTER

MULTIDIMENSIONAL GAMMA RAY SPECTROMETRY AND ITS USE IN BIOLOGY

Julian M. Nielsen and Harry A. Kornberg
Pacific Northwest Laboratories - BATTELLE-NORTHWEST
Richland, Washington

1. Introduction

Recent developments in gamma ray spectrometry [1] have made possible the identification and measurement of a large group of radionuclides in a complex mixture of radionuclides with a single measurement on the sample. These techniques have the potential for wide applicability in the field of biology especially in those areas where it is desirable or imperative to determine many radionuclides at very trace levels in complex biological samples without the complication of chemical separation. This paper describes an instrumental technique which was designed specifically for this purpose.

Accommodation of large samples is desirable in low level spectrometry so that minimum sample preparation is needed and to provide enough radionuclide to be measurable above the background with sufficient precision without impractically long counting periods. This requires large detector systems. Since this spectrometry is based on detection of gamma rays by their characteristic photoelectric interaction in the detector, the detector also must be large to favor photoelectric interaction over Compton interaction. Through absorption and anticoincidence shielding the background due to natural radionuclides and cosmic rays is reduced. The anticoincidence shielding also operates to greatly improve the selectivity (the ability to measure one radionuclide in the presence of other radionuclides) by cancelling counts due to Compton events whose degraded photons interact with the shield.

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RECEIVING SECTION.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

In multidimensional gamma ray spectrometry great additional selectivity is obtained by providing an additional parameter of separation based on the property of most radionuclides to decay through emission of two or more gamma rays in sequence (cascade). A multidimensional analyzer analyzes the cascade gamma rays received at the same time ("in coincidence") according to their energies and stores them uniquely in the memory according to the energy lost by each of them in the two primary crystals of the detector. Radionuclides which decay by single or non-coincident gamma emission do not provide coincident signals to the analyzer so their counts are stored only on the axes of the analyzer memory. This greatly improves the selectivity while effectively decreasing the background and Compton interference by orders of magnitude.

2. Instrumentation

The detector system is shown in Figure 1. It consists of two anti-coincidence shielded 4 inch thick by 6 inch diameter NaI(Tl) detectors. One of these detectors has a 4 inch light pipe of pure sodium iodide which serves to shield this crystal from the gamma radiation originating in its phototube. Although it was incorporated in this system only on a trial basis it resulted in a very worthwhile background reduction to one-half that of the crystal without the light pipe. The anticoincidence annulus is a NaI(Tl) crystal 11-1/2 inches in diameter by 12 inches long with a 6-1/2 inch diameter center hole to accommodate the principal detectors. This anticoincidence crystal need not be of spectrometry grade sodium iodide since its response is used without energy analysis only to operate the anticoincidence gate of the analyzer so that signals from the principal detectors are not accepted for analysis during the period when a signal is received from the annulus crystal. This reduces the background by eliminating from analysis cosmic ray events occurring simultaneously in the principal detector(s) and the anticoincidence shield as well as reducing the interference from those events wherein full energy absorption of the gamma ray does not occur in the principal detector and the degraded photon from the Compton event interacts with the shield crystal.

Amplified signals from the two principal detectors are routed to a 4096 channel analyzer (the analyzer used in this work was a Nuclear Data

model). The 4096 channels were used in a rectangular 64 x 64 channel grouping where signals were stored in energy groupings of units of 47 kev (e.g., channel 1 on the x axis stores pulses from crystal 1 between the energies of 0 and 47 kev, channel 2 stores pulses from 47 kev to 94 kev, channel 3 from 94 kev to 141 kev, etc.; channel 1 on the y axis stores pulses from crystal 2 between the energies of 0 and 47 kev, etc.). If a simultaneous (coincidence) event occurs in both crystals the pulse will be stored in a channel on the plane of channels bounded by the x and y axes and whose coordinates are determined by the energies of the events in the two detector crystals. Thus in the case of Co^{60} if the 1.33 Mev gamma rays were absorbed in crystal 1 and the 1.17 Mev gamma rays were absorbed in crystal 2, the events would be stored in a Gaussian distribution depending on the resolution of the detection system and centered near the channel with an x coordinate of $28 (\sim \frac{1.33}{.047})$ and a y coordinate of $25 (\sim \frac{1.17}{.047})$. Since it is equally likely that crystal 1 will get the 1.17 Mev gamma and crystal 2 the 1.33 Mev gamma a similar distribution of stored events will occur at the channel with an x coordinate of 25 and a y coordinate of 28. These two peaks can be seen in a picture from the oscilloscope display of the analyzer shown in Figure 2. Each radionuclide has a different distribution of gamma rays and hence gives a characteristic spectrum. Because of the added dimension of separation the channels in which each radionuclide are stored are separated more completely from and are much less affected by contributions from other radionuclides.

The analyzer has a photographic readout (Optikon) which permits a readout of the memory on four Polaroid prints in about four minutes. A paper tape perforator readout (Tally) is also used when data are to be analyzed by a computer. The tape unit can also be used to enter background or standard spectra data into the analyzer memory for background subtraction and spectrum stripping operations. Each channel is capable of storing 262,144 (2^{18}) events before it resets to zero and continues to store.

3. Operating Procedures and Characteristics

The entire detector assembly is housed in a shield constructed of

lead bricks to give 4 inches of lead shielding in all directions. In the shield the detector system is mounted with the cylindrical axis vertical. A geared screw mechanism operated by a hand crank on top of the lead shield is used to raise and lower the top principal detector to allow introduction of the samples. Samples as large as 6 inches in diameter and 2 inches thick have been counted. Standard spectra for all radionuclides present and background spectra for each sample shape and matrix are needed to obtain quantitative measurements.

3.1 Background, backscatter and Compton interference reduction

Many features in this counting system operate to reduce the counting background. Figure 3 shows the overall reduction obtained compared with a single crystal shielded by four inches of lead only. The background reduction with the multidimensional analysis system is three to five orders of magnitude greater than with single crystal spectrometry. The anticoincidence shield is responsible for reducing the background by about a factor of six with the major reduction due to the coincidence mode of operation.

In taking the background shown in Figure 3, a 1 inch lead absorber six inches in diameter with a center 1 inch hole for the sample was placed between the two crystals. For a background count to be stored in the multidimensional spectra (not on the axes) a gamma ray interaction (or cosmic shower, etc.) must occur simultaneously in each of the two principal detectors. The probability of this event is greatly reduced by inserting this lead absorber into the space between the detectors which is not occupied by the sample. A background reduction of a factor of four is obtained relative to the system without an absorber.

The use of a lead absorber provides an additional benefit by minimizing interference due to backscattered gamma rays. Some single gamma rays can lose part of their energy in one crystal and be scattered back to the other detector. This event could not be distinguished by this system from two cascade gamma rays of energies corresponding to the energies lost by the single gamma ray in each crystal. The lead absorber reduces this backscatter. Perkins [1] has reported that for Zn^{65} (1.117 Mev single gamma) the lead absorber reduces the backscatter by at least a factor of 8. Such a reduction is especially desirable in measurement of most biological

samples containing large amounts of natural K^{40} , which decays with single 1.47 Mev gamma ray emission.

Compton events are mostly cancelled by the anticoincidence annulus crystal. Some are counted but their interference with measurement of the photopeaks of other radionuclides is much less than with single crystal spectrometry. Because of the added dimension the Compton events occur mainly in bands a few channels wide extending from each axis to each photopeak. Photopeaks from other radionuclides generally fall in areas little affected by these Compton areas.

3.2 Counting Efficiency and Sensitivity

For highest efficiency counting a total absorption detector is needed. For gamma rays of about 1 Mev this would require a sodium iodide detector at least 6 inches thick in each direction from the sample. The system described here represents a compromise in order to utilize the largest NaI(Tl) anticoincidence annulus we could obtain. Large crystals are especially needed for high efficiency coincidence counting where the efficiency is two times the product of the efficiencies of the individual crystals (with a small correction for angular correlation factors for the individual radionuclides). Thus, for example, the dual 6 inch diameter by 4 inch thick crystals have an efficiency for a 1 Mev single gamma emitting of 38%. When used in the coincidence mode for counting two 1 Mev gamma rays in cascade the efficiency is 7.2%. The same values for dual 3 inch diameter by 3 inch thick crystals would be 12.5% and 0.78%. Thus in this case the larger crystals have an efficiency for coincidence counting greater than the standard 3 inch by 3 inch crystals by a factor of 9. Where interaction of three gamma rays is involved in the counting (as with Na^{22} , Ag^{110m} , Sb^{124} and Ba^{140}) the factor is considerably larger in favor of the larger crystals.

High sensitivity results from many factors, mainly the high efficiency, low background, minimum Compton and background interference, and great selectivity. As a typical example Sc^{46} in a Columbia River water sample containing numerous other radionuclides can be measured at an efficiency of about 12% (or 6% with the 1 inch lead absorber). The background with the absorber is about 1 count per hour. A sample containing 2 dpm would give 120 counts in 1,000 minutes above a background of 15 counts.

4. Applications To Biological Studies

In some ways biological materials are especially suited to measurement by gamma spectrometry. Because living matter has a relative low density and consists mainly of atoms of low atomic number, self-absorption of gamma rays by the sample material itself is low even with large sample sizes. This simplifies detector calibration, gives low Compton interference, and allows sample size reduction by ashing, etc. In using gamma spectrometry for trace element analysis on biological samples which have been neutron activated it is advantageous that hydrogen, carbon, nitrogen and oxygen which constitute 96% of living matter do not produce interfering radionuclides. In both modern radioecological and trace element studies there are usually present in the sample to be measured a large number of radionuclides. Since most radionuclide counting techniques have been unable to determine many radionuclides directly on the same sample, radiochemical separation has been necessary. Multidimensional gamma ray spectrometry has greatly improved selectivity which allows many more radionuclides to be determined by direct measurement without prior chemical separation.

4.1 Environmental Radionuclide Measurement

In Table I are given analytical data obtained by multidimensional gamma ray spectrometry on some typical environmental samples. These samples were measured without chemical separation. The water and urine samples were boiled down to the salts. The clams were counted without treatment. The catfish and shiner sample was dried and the caribou sample was ashed. The air sample was a membrane filter pressed into a small disc for counting. The dashes given in the table indicate that this radionuclide could be determined by this technique but was not measured on this particular sample. In some cases a sample was measured twice at different times to allow decay of some of the shorter lived radionuclides with a subsequent increase in precision of measurement of the remaining longer lived radionuclides.

These data indicate the feasibility and ease of radioecological studies using the multiple tracers available in the Columbia River water from reactor operations and in the air in Alaska from stratospheric bomb fallout. With this technique above 18 radionuclides can be measured in the river water.

Most of these can also be determined in biota in the river such as the fish and the clams reported here. In addition, 9 of the radionuclides can be measured in the urine of humans using this river water. Similarly, 13 radionuclides can be measured directly by this technique in the air in Alaska from stratospheric fallout or cosmic ray spallation [2]. These radionuclides are taken up by the lichens which are an important food source for Alaskan caribou. The radionuclides in the air can be measured in the lichens, and 8 of them can be determined in the liver of caribou eating these lichens.

4.2 Trace Element Measurement by Neutron Activation Analysis

An important application of this counting technique when combined with neutron activation analysis is in the study of certain trace elements in the blood. In order to illustrate the use of this technique without chemical separation only those elements are reported in Table II which are directly measurable using one 1000 minute count after the Na^{24} and Br^{82} have decayed to low levels. The Na^{24} can be measured at any time for about two weeks and Br^{82} can be measured after three weeks. These measurements are not reported here as trace elements. To obtain the data in Table II 100 microliter samples of blood or blood fraction were dried in quartz capsules and irradiated for one week in a neutron flux of about 5×10^{13} n/cm²/sec. Four weeks were allowed for decay of the shorter lived radionuclides. The sample was then dissolved and plated on a lucite disc 1/4 inch thick and 1 inch in diameter. A duplicate second disc mounted on top completes the sample assembly which was designed to minimize the amount of bremsstrahlung radiation. Measurements were then made by multidimensional gamma ray spectrometry of Sc^{46} , Fe^{59} , Co^{60} , Zn^{65} , Ag^{110m} , Sb^{124} , and Cs^{134} . From these measurements and those obtained from appropriate standards similarly treated, the trace element concentrations were calculated and are reported in Table II.

Although these data represent only single samples for each species, several interesting implications may be drawn. Most striking is the relatively high value for cobalt in human serum. This may have been due to individual variation since cobalt in human whole blood has been reported at 0.3 $\mu\text{g/g}$ [3]. Another possibility is that dietary intake of enriched foods, as are now fairly ubiquitous in the U.S.A., is causing gradual increases in trace elements associated with vitamins. No value for scandium in blood serum was found in the literature. However, whole blood is reported [3] to contain 600 times as much as we found. That this difference is probably not due to different scandium concentrations in serum and whole blood is suggested by comparing scandium values we found in swine serum and whole blood.

Some interesting differences are apparent when swine serum, plasma, whole blood, and cell lysate values are compared for several of the trace elements. (The difference between serum and plasma was the presence of heparin in the latter, and the difference between whole blood and cell lysate is the presence of distilled water in the latter.) It appears that the heparin used contained scandium, silver, and antimony, with possibly also cobalt and zinc being present. Similarly, it appears that the distilled water used to lyse the cells contained silver and antimony. Unfortunately, separate analyses were not performed on the heparin and distilled water. However this was a preliminary experiment designed to test methodology, rather than to obtain definitive values.

For the most part, no striking differences in trace element concentrations were found among the different species. Iron and zinc values found agree with those reported for humans in the literature [4,5]. The level of silver we measured in humans is lower than that reported by Gofman [4] by a factor of 140. The antimony value we found for human blood serum agrees with that reported by Bowen and Cawse for whole blood [3]. Cobalt values for the rat, dog, goat, sheep, and swine generally agree with values reported in the literature for humans [3,5]. Cesium values we found in serum of the several species are lower than those reported by Gofman [4] for human serum by factors of about 400, but agree fairly well with the reported value for whole blood. We were unable to find any reports in the literature of scandium, silver, or antimony in serum.

In this study multidimensional gamma ray spectrometry allowed precise measurements of trace elements in blood and blood fractions, some of which had not been measured before. The special advantages of requiring no chemical separation to determine seven elements in one sample greatly simplified the study and eliminated the sources of much possible contamination.

The results reported here only serve to indicate the wide range of applicability of multidimensional gamma ray spectrometry.

REFERENCES

- [1] PERKINS, R.W., Nuclear Instruments and Methods, in press, (1965).
- [2] PERKINS, R.W., NIELSEN, J.M., and THOMAS C.W., Science 146 (1964) 762/764.
- [3] BOWEN, H.J.M., and CAWSE, P.A., U.K.A.E.A. Report AERE-R 4309 (1963).
- [4] GOFMAN, J.W., in Advances in Biological and Medical Physics VIII (TOBIAS, C.A., LAWRENCE, J.H., and HAYES, T.L., eds.) Academic Press Inc., New York (1962) 1-39.
- [5] LUCKEY, T.D. and COHAGEN, D.L., in Blood and Other Biological Fluids (ALTMAN, P.C. and DITTMER, D.S., eds.) Federation of American Societies for Experimental Biology, Washington, D.C. (1961) 21-23.

TABLE I
SOME TYPICAL EXAMPLES OF CAPABILITY OF MULTIDIMENSIONAL
SPECTROMETRY FOR ANALYSIS OF ENVIRONMENTAL SAMPLES

Radio- nuclide	Columbia River dpm/l	Catfish & Shiners dpm/kg	Clams dpm/g	Human Urine dpm/l	Air, Alaskan dpm/10 ⁶ ft ³	Liver, Caribou, Alaskan dpm/kg
Be ⁷					2,820	
Na ²²	<0.02	0.83		1.5	2.2	.783
Na ²⁴	3,050	--	--	20		
Sc ⁴⁶	45	12	70	2		
Cr ⁵¹	21,500	--	1,360			
Mn ⁵⁴	15.6	15	11		189	320
Mn ⁵⁶	--	--	1,520			
Co ⁵⁸	25	4.5	54			
Fe ⁵⁹	12.5					
Co ⁶⁰	4.6	105	13	0.06	2.1	85
Cu ⁶⁴	1,520	--	--	2		
Zn ⁶⁵	510	26,300	254	12		87
As ⁷⁶	500		19.4	40		
Y ⁸⁸					1.4	
Zr ⁹⁵ -Nb ⁹⁵	9.4				192	
Ru ¹⁰⁶	8		8		885	
Ag ^{110m}						461
Sb ¹²⁴	27	<100	2.3		1.9	
Sb ¹²⁵					350	
Cs ¹³⁴	0.05	11		0.6	0.50	36
Cs ¹³⁷	1	190		150	422	23,000
Ba ¹⁴⁰	31	865	12		55	
Ce ¹⁴⁴					1,770	
Th ²³²						10

TABLE II
TRACE ELEMENT STUDY DATA OBTAINED BY NEUTRON ACTIVATION AND
MULTIDIMENSIONAL GAMMA RAY SPECTROMETRY WITHOUT CHEMICAL SEPARATION

Sample	Element Concentration (µg/g)						
	Sc	Fe	Co	Zn	Ag	Sb	Cs
Serum, Human	0.042	1,050	25	798	1.44	1.5	0.50
Serum, Rat	0.019	7,000	1.0	1,990	11.0	12.6	0.58
Serum, Dog	--	3,010	1.1	1,350	0.21	0.49	--
Serum, Goat	0.028	2,240	1.1	835	0.52	19.5	0.34
Serum, Sheep	0.007	2,750	0.9	1,150	0.58	0.25	0.36
Serum, Palouse Swine	0.020	2,470	1.5	965	3.6	0.82	0.75
Serum, Miniature Swine	0.013	2,890	1.6	1,020	1.4	0.88	0.81
Serum, Miniature Swine	0.009	2,810	1.7	875	3.2	1.7	0.73
Plasma, Miniature Swine	0.057	2,730	2.3	1,150	9.2	3.8	0.65
Whole Blood, Miniature Swine	<0.04	427,000	1.4	5,560	2.7	2.3	3.3
Cell Lysate, Miniature Swine	<0.04	403,000	0.8	4,930	12.7	9.5	2.9

FIGURE 1

DETECTION SYSTEM FOR MULTIDIMENSIONAL GAMMA RAY SPECTROMETER

DUAL CRYSTAL COINCIDENCE MULTIDIMENSIONAL GAMMA RAY SPECTROMETRY

SINGLE CRYSTAL GAMMA RAY SPECTROMETRY

FIGURE 2
COMPARISON OF SINGLE CRYSTAL AND
MULTIDIMENSIONAL GAMMA RAY SPECTRA

FIGURE 3
COMPARISON OF THE BACKGROUND COUNTING RATES OF
A SINGLE CRYSTAL WITH THAT OF THE TWO CRYSTAL
MULTIDIMENSIONAL DETECTOR SYSTEM.