MAY 4 1965

BNWL-SA-137
DNRA-137

CONF-650405-5

SELECTIVE AND SENSITIVE ANALYSIS OF ACTIVATION PRODUCTS
BY MULTIDIMENSIONAL GAMMA RAY SPECTROMETRY

By

R. W. Perkins* & D. E. Robertson*

Chemical Research
Chemistry Department
BATTELLE-NORTHWEST

MASTER

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. No United States nor the Commission, nor any person acting on behalf of the Commission, 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, to the extent that such 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.

ABSTRACT

Gamma-ray spectrometry detector systems have been developed for use with multidimensional analyzers which permit an extremely sensitive and selective analysis of complex radionuclide mixtures. With these systems, use is made of the individual gamma-ray decay characteristic of each radionuclide for identification and measurement. This is accomplished by sandwiching the sample between two detectors and canceling those events not totally absorbed in these detectors with a third annular detector which serves as an anti-coincidence guard. The sample can be held in a fourth annular disc shaped detector which allows the sample to be seen by the two principal detectors but which intercepts and cancels most of the degraded photons which would be scattered between them. The principal detectors are shielded from the gamma-ray radiation originating in their phototubes with light pipes which provide a substantial background reduction.

Applications of these instruments include the direct measurement of atomic-weapons produced radionuclide and cosmic-ray produced activation products in the atmosphere and the direct measurement of neutron activation products in water, soil and biological materials.

1. Introduction

A common problem both in activation analysis and in various radiochemical studies is the measurement of several constituents in a complex mixture of radionuclides. These radionuclide mixtures may be intentionally produced for activation analysis or they may be the result of nuclear weapons testing, nuclear reactor and chemical processing plant operations, emanation gases from natural radionuclides in the earth crust, cosmic ray spallation reactions in the atmosphere, or other sources. The sample compositions vary widely in both absolute and relative concentrations and their analysis

*General Electric Company, Richland, Washington. Work performed under Contract No. AT(45-1)-1350 for the U. S. Atomic Energy Commission. Now work is performed for Battelle-Northwest under Contract No. AT(45-1)-1830.

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.

is often extremely difficult. In recent years special gamma-ray spectrometry coincidence techniques⁽¹⁾ and anticoincidence shielding methods⁽²⁾ have greatly simplified the measurement of many radionuclides where they are present as major constituents. Through our recent developments in multidimensional gamma-ray spectrometry⁽³⁾ it is now possible to perform direct instrumental analysis of numerous complex radionuclide mixtures^(3,4,5,6,7,8) which previously required both extensive radiochemical separations and very sensitive counting techniques. The selectivity provided by this technique allows the measurement of many radionuclides at orders of magnitude lower concentration than that of the sample's major constituents; however, these measurements do require long counting periods which usually range from 30 to 1000 minutes.

In this present work a new multidimensional detector system is reported and compared with our original detector system which was described earlier for multidimensional gamma-ray spectrometry⁽³⁾. Both of these detector systems provide ultra-low backgrounds and high counting efficiencies plus a low Compton response and at the same time use coincidence techniques to make use of the individual gamma-ray decay characteristic of each radionuclide for its identification and measurement. In these spectrometer systems the sample is usually viewed by two detectors for normal and coincidence gamma-ray spectrometry and these principal detectors are contained in a third detector which serves as an anticoincidence annulus for Compton and background suppression. The new detector system is designed to further minimize Compton and backscatter response, and it employs large plastic phosphors rather than sodium iodide crystals for anticoincidence shielding.

2. Instrumentation

Design of Detector Systems

In the quantitative measurement of radionuclides in a complex radionuclide mixture the Compton events drastically reduce both the precision and sensitivity of the measurement of many radionuclides. Much of this Compton interference can be removed by anticoincidence shielding techniques⁽²⁾ which involve surrounding the principal detector, as nearly as practical, with a second detector in anticoincidence with it. This technique also reduces the normal backgrounds of NaI(Tl) detectors by about a factor of 4 in the energy range from 0 to 3 Mev and has been incorporated into the design of both multidimensional gamma-ray detector systems.

The original multidimensional gamma-ray spectrometry system used a 4096 channel analyzer in a 64×64 channel grouping arrangement in conjunction with two anticoincidence shielded 4 inch thick by 6 inch diameter NaI(Tl) detectors (see Figure 1). One of the 4 inch by 6 inch detectors had a 4 inch light pipe of pure sodium iodide which served to shield the detector from gamma radiation originating in its phototube. The anticoincidence annulus was a 11-1/2 inch diameter by 12 inch long NaI(Tl) crystal with a 6-1/2 inch diameter center hole to accommodate the principal detectors. The crystal cladding was 19 mil stainless steel and thus limited the effectiveness of the anticoincidence shield for Compton suppression.

The new detector system is shown in Figure 2. The two principal detectors are 5 inches in diameter by 6 inches thick NaI(Tl) crystals and have 8 inch long lucite light pipes between the crystals and their phototubes. These light pipes serve to shield the crystals from their phototubes but also shield them from external radiation which could reach the crystals without traversing the anticoincidence shield. The resolutions of these crystals for the Cs¹³⁷ line with a point source centered on top were just under 10%. Without the light pipes their resolutions would have been better than 9%. The crystals are each enclosed in a 30 inch diameter by 15 inch thick plastic phosphor which serves as an anticoincidence shield (see Figure 2). The bottom plastic phosphor is viewed with six 5 inch diameter EMI phototubes (No. 9530B) while the top phosphor is viewed with four 7 inch diameter EMI phototubes (No. 9623B). Surfaces of these phosphors which are not viewed by the phototubes are covered with a white reflective coating (Pilot reflective coating) and are made light tight by wrapping them with 3 or 4 layers of 1 mil aluminum foil. The top plastic phosphor is raised and lowered on a lifting jack which is driven with a variable speed motor. The top NaI(Tl) crystal can be raised and lowered independently with a mechanical lifting device which is mounted on the top plate of the plastic phosphor. An optional detector in this multiple detector system is an annular disc shaped NaI(Tl) crystal which can be located between the two principal NaI(Tl) detectors and covers the area not occupied by the sample. This detector is 6 inches in diameter by 1.5 inches thick with a 1.5 inch diameter center hole. It is viewed through four 1 inch diameter by 6 inch long lucite light pipes with 1-1/8 inch diameter phototubes (EMI 9524B) and the outputs from these tubes are added to the coincidence output from the anticoincidence shield. This detector serves to minimize storage

by the analyzer of backscatter events between the crystals. The resolution of this detector for the 0.662 Mev line of Cs^{137} was 22%. The cladding of all of the crystals and inside cladding on the phosphors have been kept to a minimum to reduce the number of interactions with inert material in the detector system. The faces of the annular NaI(Tl) crystal are 15 mil aluminum and no light reflective material is located between this aluminum and the crystal faces. The principal NaI(Tl) crystals are also clad in a 15 mil aluminum and a minimum of light reflecting material is used between this cladding and the crystals.

The all sodium iodide system is presently operated with a Nuclear Data 4096 Channel Analyzer while the new detector system is operated with a Packard 4096 Channel Analyzer. Both of these analyzers are equipped with fast printers which permits a readout of the entire memory in about four minutes. Tally paper tape units are used as a readout for computer calculation. The sample to be measured is sandwiched between the principal detectors (or is located in the center of the annular detector). The amplified signals from the two principal detectors are fed to the two separate analog to digital converters of the analyzer for energy and coincidence analysis and from there to the memory storage unit. When a single photon interacts with one of the detectors, the event is stored on the corresponding X or the Y axis of the memory. When two photons are emitted simultaneously, each interacting with a separate detector, the event is stored in an energy-energy plane at a point uniquely characteristic of the two energies. In the new detector system, gamma rays which are not totally absorbed by one of the two principal detectors have a high probability of interacting with the plastic phosphors or the annual backscatter NaI(Tl) crystal which results in the event being canceled. The spectrum of Co^{60} illustrates (Figure 3) the manner of storage. The normal spectrum of Co^{60} as seen by one detector located outside of the anticoincidence shield is shown for comparison. With detection systems of these types the unique decay characteristic of many radionuclides are readily resolvable thus permitting a far greater sensitivity for detection and direct measurement of many minor radioactive constituents.

Background, Backscatter, and Compton Response

The background reduction which can be achieved by anticoincidence shielding for a single crystal in the all sodium iodide system is shown in Figure 4. The top curve is the normal crystal background. The next curve down is the

background obtained with the 4 inch long pure NaI light pipe between the crystal and its phototube and represents about a 20-50% background reduction. The third curve down and the bottom curve are background measurements of this crystal with its associated light pipe placed inside the annulus and with a second 6 inch diameter by 4 inch thick crystal above it as shown in Figure 4. It is evident that the background is reduced somewhat simply by the NaI shielding in this arrangement. When the output pulses from the annulus and top crystal are connected in anticoincidence with the principal detector a very substantial total reduction is obtained ranging from a factor of 5 to 10 except at the K^{40} peak. The fourth curve down is the background for the top 6 inch diameter by 4 inch thick NaI(Tl) crystal with the annulus and bottom crystal operating in anticoincidence with it. The background of this crystal is 2 to 3 times higher than that of the bottom crystal and this is the result of these factors: (1) It is located at the top rather than the center of the anticoincidence annulus and it is not shielded as well as the bottom detector from external gamma-rays and particularly cosmic ray showers; (2) It does not have the NaI(Tl) light pipe to protect it from radiation originating in its phototubes. It should be pointed out that the background reduction of 20-50% obtained by use of the light pipe (upper two curves) is extremely important if one is trying to obtain maximum background reduction by anticoincidence shielding. This fraction of the original background is due to the phototube and is not effectively removed by anticoincidence.

In Figure 5 the backgrounds of the single bottom crystals from the all sodium detector system and from the new plastic phosphor detector system are shown. The normal backgrounds at the top of the graph are simply those obtained with the detectors removed from their anticoincidence shields and counted in a separate 4 inch thick lead shield. The anticoincidence background shown in the bottom curves were obtained during normal operation of the multiple detector systems with the two principal detectors together. These backgrounds are therefore those of the bottom crystals while the top crystals as well as the anticoincidence shields were operating in anticoincidence with them. It is evident that while there is little difference between the normal backgrounds of these crystals, the anticoincidence backgrounds are considerably different. The combined effect of this very large plastic anticoincidence shield plus the longer light pipe provides this system with better shielding and a lower background from external gamma radiation; however, a large photopeak at 2.225 Mev is produced which is due to cosmic ray interactions with this plastic. This 2.225 Mev gamma-ray is due to the reaction

of cosmic ray produced neutrons, which are produced both in the lead and plastic shielding, with hydrogen atoms in the plastic, $H^1(n,\gamma)H^2$. This effect has been reported previously both during use of plastic phosphor^(9,10) and paraffin shielding⁽¹¹⁾. This gamma ray does not produce a significant increase in the energy-energy plane background; however, it is undesirable and means of reducing or eliminating it are being considered.

To store a background count in the energy-energy plane, a gamma-ray interaction (or cosmic-ray shower, etc.) must occur in each of the two principal detectors simultaneously. The probability of such an event occurring can be greatly reduced by covering the area between the detectors which is not occupied by the sample with a lead absorber. Such an arrangement provides an additional benefit in minimizing backscatter between these detectors as will be pointed out, but it has the obvious disadvantage of decreasing the geometry and providing a large mass of inert material with which the gamma-rays from the sample can interact. For the all sodium iodide crystal system, 6-3/8 inch diameter lead absorbers of 1/2 or 1 inch thickness with center holes the size of the samples (1 to 2 inches in diameter) have been used. The effect on the background in the energy-energy plane of using these absorbers, and the anticoincidence shielding, for the all NaI(Tl) system is shown in Figure 6. This figure presents curves of the background counting rate on an energy diagonal extending from 0 to 3 Mev in the energy-energy plane. The top three curves show the backgrounds obtained with no absorber, and with a 1/2 inch and 1 inch thick lead absorber (with 1 inch diameter center holes) between the crystals. The crystal spacing was increased to 1/2 inch and to 1 inch respectively to accommodate these absorbers; however, it has been observed that increasing the crystal spacing from 0 to 1 inch only reduces the background by about 25%. The background reductions relative to the no-absorber curve average about a factor of 2 and 4 respectively for the 1/2 inch and 1 inch absorbers. In the anticoincidence mode of operation these backgrounds are reduced by an additional factor of 5 to 20. If one selects a 1 inch thick absorber the total decrease in background over that obtained without the absorber and without anticoincidence ranges from a factor of 20 to 50. Backgrounds in other areas of the energy-energy plane are reduced in approximately proportional amounts by this combination of absorbers and anticoincidence shielding⁽³⁾.

Figure 7 compares the backgrounds of the all sodium iodide system and the

plastic phosphor shielded system for the energy diagonal from 0 to 3 Mev. These upper backgrounds are for the systems with their principal detectors together while the lower backgrounds were measured with the 1 inch thick lead absorbers between the crystals. The combined effect of having light pipes on both of the principal detectors and the larger plastic phosphor anticoincidence shield of the new detector system provides a 2 to 10-fold lower background over most of the energy-energy plane for both cases. These curves are based on very low counting rates and indicate the magnitude of background variation rather than the precise background response in these energy areas.

In Figure 8 the background of the single 4 inch thick by 6 inch diameter crystal without lightpipe or anticoincidence is compared with that of the two detectors in the multidimensional detector system separated by a 1 inch thick lead absorber. The background shown for the multidimensional detector system is for an energy diagonal from 0 to 3 Mev and therefore represents the case of 0 to 3 Mev absorption by each detector. These curves can also be used to compare the background that would be obtained with two crystals used in a summing arrangement with that shown for the two crystals in the multidimensional arrangement. In this case the background at a given energy on the top curve should be doubled and compared with a point of half that energy on the bottom curve. By either this method of comparison or that shown in Figure 8, the background with multidimensional analysis is three to five orders of magnitude lower. With the new detector system using the 5 inch by 6 inch crystals this background is an additional factor of 2 to 10 lower yet (see Figure 7).

One of the problems in using high geometry detector arrangements such as these is that some of the single gamma rays which interact with one of the detectors are backscattered and absorbed by the second crystal thus producing a coincidence pulse which is stored in the energy-energy plane. This effect is, of course, minimized in some areas by the use of the lead absorbers. Figure 9 shows backscatter curves of Zn^{65} , (as measured with the all sodium iodide system) in the energy-energy plane on a constant energy line connecting the X and Y axes. These curves are for a point source with no absorber and with the 1/2 inch and 1 inch thick absorbers with 1 inch diameter center holes. The curves are normalized to a counting rate of 10,000 counts in the peak channels on the X and Y axes. It is evident that the absorbers reduce backscatter by factors of 8 to 50. This fact becomes especially important in the measurement of trace constituents in biological materials where large

amounts of natural K^{40} are present. This radionuclide decays by emission of a single 1.47 Mev gamma ray and its contribution in the energy-energy plane due to backscatter is a problem. Figure 10 shows the backscatter curves for a point source of Co^{60} measured in a similar manner. The constant energy line in this case connects the two additional peaks at 2.5 Mev on the X and Y axes and shows the two coincidence photopeaks (used for Co^{60} measurement) in the energy-energy plane. The X and Y axes counting rates are again normalized to 10,000 counts and the curves show the large backscatter reduction affected by the lead absorbers. A disadvantage of the lead absorbers is that the energy lost to the crystal before backscatter into the lead absorber is recorded by the analyzer. Part of this is stored on the X or Y axes but part is also stored on energy lines which leaves the axes at a right angle and connect with the photopeaks in the energy-energy plane. In addition, some gamma rays interact with the lead absorbers and are then scattered into the detectors. To eliminate these problems the use of a NaI(Tl) detector for an absorber which serves as part of the anticoincidence shield was studied.

In Figure 11 the energy-energy response of the new detector system to Co^{60} gamma-rays is compared for different absorber materials. As shown in this illustration the detectors were positioned 1.5 inches apart, and a point source of Co^{60} was centered between them in their 1.5 inch diameter center hole. The relative counting rates on the "total absorption line" (exclusive of the photopeaks) and on "Compton lines" are compared for absorbers of air, NaI(Tl), NaI(Tl) in anticoincidence and lead. For ease in comparison the counting rates on these lines relative to their Co^{60} photopeaks have been normalized to unity for the air absorber. The effects of the NaI(Tl) crystal (with and without anticoincidence) and of the lead absorbers on the total absorption line are similar, reducing the total counting rates to 0.2 to 0.084 of that observed with an air absorber. The effect of these absorbers on the Compton lines are considerably different. While the lead absorber and the NaI(Tl) actually increase the backscatter lines by more than a factor of two, on operating the NaI(Tl) crystal in anticoincidence these lines are decreased. In addition, with the NaI(Tl) absorber in anticoincidence the Compton contribution is lowered throughout other positions of the energy-energy plane and is 5 to 20-fold lower than with a lead absorber on the X and Y axes.

A detailed comparison of the Compton response of the plastic phosphor shielded system with the all sodium iodide system is not included in this paper; however, the Compton response of the new system is much lower. With the crystal

faces together in each system (and no absorbers) the Compton response both on the X and Y axes and in the energy-energy plane is 2 to 4-fold lower over most of the energy regions of interest. This is due both to the thinner cladding of the new system and the better approach to 4π anti-coincidence shielding which is accomplished by use of the larger plastic anticoincidence shield.

Counting Efficiency

The sensitivity for the measurement of a given radionuclide is dependent on the instrument's counting efficiency as well as its background counting rate and the Compton interference from other radionuclides. In using coincidence counting where high efficiency is required it is essential to have rather large crystals which view the source with a good geometry since the coincidence counting efficiency varies with the square of the single crystal counting efficiency. For this discussion the counting efficiency (E) for a given gamma-ray energy is defined as the fraction of the gamma-rays emitted by a source that are totally absorbed by one of the two principal detectors. Since the sources are positioned symmetrically between two detectors, the total counting efficiency would be $2E$.

Where two gamma-rays are emitted in cascade, the probability for total absorption of both gamma rays in one detector or of a specific gamma ray in one and the second gamma ray in the other detector is approximately $E_1 E_2$ (neglecting the small angular correlation factor). The total coincidence counting efficiency which is here defined as the probability of total absorption of one gamma ray (either gamma ray in this case) in each of the two detectors is therefore approximately equal to $2E_1 E_2$. This factor gives the probability that a coincidence event in the sample will be stored in a total absorption peak (either one of the two) in the energy-energy plane. To illustrate the approximate efficiencies of the detector systems considered thus far, and for a larger and small system, their total counting efficiency factors ($2E$) and the total coincidence counting efficiency factors ($2E_1 E_2$) are plotted as a function of energy in Figure 12. Curves of $2E_1 E_2$ are plotted for the special case where the two gamma rays have equal energy but they do serve to illustrate the change in efficiency with energy. The approximate efficiency for the measurement of any cascade can be obtained from the equation:

$$\text{Total Coincidence Counting Efficiency} \approx 2E_1 E_2$$

where the E values can be obtained from Figure 12 (by dividing the $2E$ values by 2). The exact equation would contain a small correction factor for the

angular correlation of the gamma-ray cascade in question. The coincidence efficiency curves show the large dependence of crystal size on counting efficiency. This difference is further accentuated as the crystals are moved apart. The use of crystals smaller than 5 inch diameter would be of very limited value in multidimensional gamma-ray spectrometry. The relatively low counting efficiency of the 9-3/8 inch diameter by 4 inch thick crystal at low energies is due to its thick (19 mil) stainless steel container and to the thick coating of light reflecting material between the crystal and its container. The coincidence counting efficiencies which can be determined from Figure 12 are for thin sources sandwiched between two crystals and in the case of the 6 inch diameter detectors these are reduced by approximately 50 to 60% when the source is counted in the 1 inch diameter hole of a 1 inch thick lead absorber. The coincidence counting efficiencies which can be determined for 5 inch diameter systems (in Figure 12) are reduced by about 65% when the geometry shown in Figure 11 is used.

3. Applications

These instruments have proven to be extremely useful in the quantitative analysis of numerous complex radionuclide mixtures. A few of these applications are summarized below.

Measurement of Airborne Radionuclides

At the present time it is possible to make direct measurement of thirteen radionuclides on air filter samples^(4,12). These include Be⁷, Na²², Mn⁵⁴, Co⁶⁰, Y⁸⁸, Zr⁹⁵-Nb⁹⁵, Ru¹⁰⁶, Ag^{110m}, Sb¹²⁴, Sb¹²⁵, Cs¹³⁴, Cs¹³⁷, and Ce¹⁴⁴. The Be⁷ and Na²² are produced continuously by cosmic ray spallation reactions in the atmosphere while the other radionuclides have resulted from nuclear weapons testing. The radionuclides Na²², Co⁶⁰, Y⁸⁸, Ru¹⁰⁶, Ag^{110m}, Sb¹²⁴, Cs¹³⁴, and Ce¹⁴⁴ are measured from the counting rates of their characteristic coincidence gamma-ray energies. The energy areas used in their measurement are indicated in Figure 13. The remaining 5 radionuclides are measured on the X and Y axes after appropriate corrections for the Compton contribution from radionuclides which are precisely measured in the energy-energy plane.

Discovery and Measurement of Cosmic Ray Produced Radionuclides

The cosmic-ray produced radionuclides S³⁸ and Cl³⁸ were observed for the first time⁽⁸⁾ in rain water by measurements with our multidimensional gamma-ray spectrometer. These radionuclides are produced in the atmosphere by the cosmic-ray spallation of argon. The radionuclide Cl³⁸ (37.3 min) is

particularly suited for multidimensional analysis since its decay involves the emission of two high energy photons in cascade (1.6 and 2.16 Mev). The radionuclide S^{38} (2.9 hr) decays to Cl^{38} and its concentration can therefore also be calculated from the Cl^{38} coincidence photopeak (after the initially present Cl^{38} has decayed if no chemical separation is made). In addition to these radionuclides the cosmic ray produced Cl^{39} (55 min) is readily measured (if the chlorides are chemically separated by a $AgCl$ precipitation) by its 0.246 and 1.266 Mev coincidence gamma rays. The concentrations of these radionuclides and of the cosmic-ray produced Na^{24} (15 hr) and Na^{22} (2.1 yr) are of interest in washout studies and can readily be measured in a few liters of rain water^(8,5). The energy areas used in these measurements are shown in Figure 14.

Trace Elements in Biological Materials

The concentrations of several trace elements in biological samples are presently being studied by the direct counting of daughter radionuclides following neutron activation⁽¹³⁾. The direct instrumental analysis of trace elements in biological materials following neutron activation is particularly difficult because of the large amounts of Na^{24} , Br^{82} , P^{32} , S^{35} , and Fe^{59} which are produced. Our general procedure involves a one week irradiation of 100 microliters of blood or urine (or an equivalent weight of tissue) at a neutron flux of 10^{13} or greater, followed by a two week period to allow the Na^{24} to decay. At this time the sample, or an aliquot of it, is dried on a 1/4 inch thick by 1 inch diameter lucite disc and a second disc placed over the top. This technique greatly reduces the amount of Bremsstrahlung radiation from the sample. From an initial count at this time the Br can be estimated from its Br^{82} (35.7 hr) daughter, and then after allowing its decay the seven other elements Sc, Fe, Co, Zn, Ag, Sb, and Cs can be estimated from the concentrations of their daughters Sc^{46} , Fe^{59} , Co^{60} , Zn^{65} , Ag^{110m} , Sb^{124} , and Cs^{134} . The energy areas used in the measurement of these daughter radionuclides are shown in Figure 15.

Trace Elements in Soils

Initial studies of soils have been concerned with the trace element composition of Columbia River bottom sediments. Following a short neutron irradiation and a delay of two weeks or more to allow the Na^{24} to decay, the six elements Sc, Fe, Co, Zn, Sb, and Cs can be estimated from the daughter radionuclides Sc^{46} , Fe^{59} , Co^{60} , Zn^{65} , Sb^{124} , and Cs^{134} . The energy areas used in their measurements are shown in Figure 16.

Columbia River Radionuclides

The cooling of the Hanford Reactors (at Richland, Washington) with Columbia River water results in the addition of several activation products at extremely low concentrations (a few dpm per liter or less) to the river. A study of the behavior of eleven of these radionuclides, during their travel between Hanford and the ocean, was performed by direct measurements with a multidimensional gamma-ray spectrometer⁽¹⁴⁾. Measurements included the direct counting of river water salts and of membrane filters through which the water was filtered. The energy areas used in the measurement of this group of radionuclides are shown in Figure 17.

Other Applications

Other applications to which multidimensional gamma-ray spectrometry is being applied includes the movement of natural and artificial radionuclides in the environment and through the terrestrial and aquatic food chains, and also the behavior of trace elements and of radionuclides in the atmosphere and ocean.

4. Discussion

The response of a new design in a detector system for multidimensional gamma ray spectrometry has been studied and compared with a previously reported detector system. The advantages of the new system include both lower background and lower Compton response. In addition, a backscatter canceling crystal can be used in this system to eliminate much of the detector systems backscatter response. A 30 inch diameter by 30 inch thick plastic phosphor was used as an anticoincidence shield. This size of shield was selected on the basis of the detection efficiency of plastic phosphors⁽¹⁵⁾ and previous experience⁽²⁾ in their application.

Although not mentioned in the paper both of the systems can be used for triple coincidence spectrometry using the outputs of the three sodium iodide detectors in each case. The counting efficiency of a two detector system for coincidence counting varies with the product of the efficiencies for individual gamma ray detection and if good sensitivity is required, relatively large crystals must be used. Even though large amounts of radioactivity may be available for analysis the amount which can be used is limited by the counting rate which the instrument can accept, and the measurement of low abundance radionuclides in a mixture becomes impractical if their detection efficiency is too low.

Numerous applications of this analytical technique are being developed and applied in this rapid growing field to research problems at Battelle - Northwest. These techniques are allowing rapid progress in many problems which previously required slow, tedious, and uncertain radiochemical separations.

REFERENCES

1. Perkins, R. W., in A/Conf. 15/P/2377 (1958).
2. Perkins, R. W., Nielsen, J. M., and Diebel, R. N., Rev. Sci. Instr. 31, 1344-1349 (1960).
3. Perkins, R. W., An Anticoincidence Shielded-Multidimensional Gamma-Ray Spectrometer, Nuclear Instruments and Methods, In Press. (1965).
4. Perkins, R. W., Nielsen, J. M., and Thomas, C. W., Air Concentrations of Twelve Radionuclides from 1962 Through Mid-1964, Science, 146, 762-764 (1964).
5. Perkins, R. W., Anticoincidence Shielded-Multidimensional Gamma-Ray Spectrometry for Direct High Sensitivity Analysis of Radionuclide Mixtures, Paper, No. SM 55/33, Symposium on Radiochemical Methods of Analysis, Salzburg, Austria, October 18-23, 1964.
6. Perkins, R. W., Thomas, C. W., and Nielsen, J. M., Airborne Radionuclide Measurements and Physical Characteristic Determinations. Proceedings of the 2nd, AEC Fallout Conference, November 3-6, 1964, Washington, D.C.
7. Perkins, R. W., and Nielsen, J. M., Distribution of Na²² and Cs¹³⁴ in Fallout, Foods, and People, Nature, 205, 866 (1965).
8. Perkins, R. W., Thomas, C. W., Hill, M. W., and Nielsen, J. M., Chlorine-38 and Sulfur-38 Produced by Cosmic Radiation, Nature, 205, 790 (1965).
9. Ellett, W. H., and Brownell, G. L., Nuclear Instruments and Methods 7, 56-62, (1960).
10. McNeill, K. G., and Davis, L.D., Can. J. Phys. 40, 732-38 (1962).
11. May, H. A., Radiological Physics Division Semi-Annual Report, January through June 1962, pp. 50-65, ANL-6646 (1963).
12. Perkins, R. W., Ag^{110m} in the Atmosphere, Radiological Chemistry Annual Report for 1964, Battelle-Northwest Laboratories (In Press).
13. Perkins, R. W., Work on the Direct Measurement of Trace Elements in Biological Materials, (To be Published).
14. Perkins, R. W., Nelson, Jack L., and Haushild, W. L., Behavior and Transport of Radionuclides by the Columbia River Between Hanford and the Ocean. To be presented at the 1965 Conference on Ocean Science and Ocean Engineering, June 14-17, 1965, Washington, D.C.
15. Roulston, K. I., and Naqui, S. I. H., Nucleonics 15, 86 (October 1957).

FIGURE 1

SODIUM IODIDE DETECTOR SYSTEM FOR MULTIDIMENSIONAL
GAMMA-RAY SPECTROMETRY

FIGURE-2

**SODIUM IODIDE PLASTIC PHOSPHOR DETECTOR SYSTEM
FOR MULTIDIMENSIONAL GAMMA RAY SPECTROMETRY**

Figure 3
Co⁶⁰ SPECTRA

FIGURE 4

BACKGROUND MEASUREMENTS OF A 4 INCH THICK
BY 6 INCH DIAMETER CRYSTAL IN A 4 INCH LEAD SHIELD

FIGURE 5

COMPARISON OF SINGLE CRYSTAL BACKGROUND OUTSIDE AND INSIDE OF THEIR MULTIPLE DETECTOR SYSTEMS. ALL MEASUREMENTS WERE MADE INSIDE A 4 INCH THICK LEAD SHIELD.

Channel Numbers of Both X and Y Axes
FIGURE 6

BACKGROUND MEASUREMENTS OF THE ALL SODIUM
 IODIDE DETECTOR SYSTEM ON A DIAGONAL FROM
 0 TO 3 MEV WITH DIFFERENT COINCIDENCE AND
 ABSORBER CONDITIONS

FIGURE 7

COMPARISON OF THE COINCIDENCE BACKGROUND OF TWO
MULTIDIMENSIONAL GAMMA-RAY DETECTOR SYSTEMS
ENERGY DIAGONAL FROM 0 TO 3 Mev WITH AND WITHOUT
LEAD ABSORBERS.

FIGURE 8
COMPARISON OF THE BACKGROUND COUNTING RATES OF
A SINGLE CRYSTAL WITH THAT OF THE TWO CRYSTAL
MULTIDIMENSIONAL DETECTOR SYSTEM.

FIGURE 9

Zn⁶⁵ BACKSCATTER AT THE TOTAL ABSORPTION (1.112 MEV) ENERGY DIAGONAL IN THE ENERGY-ENERGY PLANE. COUNTS PER CHANNEL ARE NORMALIZED TO 10,000 COUNTS PER CHANNEL IN THE PEAK CHANNELS ON THE AXES.

FIGURE 10

Co^{60} BACKSCATTER AT THE TOTAL ABSORPTION (2.5 MEV) ENERGY DIAGONAL IN THE ENERGY-ENERGY PLANE, COUNTS PER CHANNEL ARE NORMALIZED TO 10,000 COUNTS PER CHANNEL IN THE TOTAL ABSORPTION PEAK CHANNELS ON THE AXES.

Absorber Material	Total Absorption Line*	Compton Lines*
Air	1	1
NaI(Tl)	0.21	2.4
NaI(Tl) Anticoincidence	0.21	0.84
Lead	0.084	2.0

*Normalized to Air

FIGURE 11
BACKSCATTER AND COMPTON RESPONSE IN THE ENERGY-ENERGY
PLANE WITH DIFFERENT ABSORBER MATERIALS

FIGURE 12

TOTAL COUNTING EFFICIENCIES AND TOTAL COINCIDENCE COUNTING EFFICIENCIES OF FOUR MULTIDIMENSIONAL GAMMA-RAY DETECTOR SYSTEMS

FIGURE 13

ENERGY AREAS USED IN THE DIRECT MEASUREMENT OF 13
AIRBORNE RADIONUCLIDES ON AIR FILTERS

FIGURE 14

COSMIC RAY PRODUCED RADIONUCLIDES. ENERGY AREAS USED IN THE DISCOVERY OF Cl^{38} AND S^{38} AND IN THE DIRECT MEASUREMENT OF Na^{22} , Na^{24} AND Cl^{39} IN RAIN WATER.

FIGURE 15

ENERGY AREAS USED IN THE DIRECT MEASUREMENT OF 7
RADIONUCLIDES IN BLOOD AND URINE SAMPLES

FIGURE 16

ENERGY AREAS USED IN THE DIRECT MEASUREMENT OF 7
RADIONUCLIDES IN SOIL (COLUMBIA RIVER SEDIMENTS) FOLLOWING
NEUTRON ACTIVATION

FIGURE 17

ENERGY AREAS USED IN THE DIRECT MEASUREMENT OF 11
RADIONUCLIDES IN COLUMBIA RIVER WATER