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NaI(Tl)-Ge(Li) COINCIDENCE GAMMA-RAY SPECTROMETRY
FOR
RADIONUCLIDE ANALYSIS OF ENVIRONMENTAL SAMPLES*

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SUMMARY

A multi-parameter NaI(Tl)-Ge(Li) coincidence gamma-ray spectrometry system has been developed for the analysis of nuclear power industry radioactive effluents in environmental samples. Several detector configurations have been evaluated.

Data acquisition is by buffer storage of the ADC output and final recording of each event in list mode on magnetic tape. A 16K computer and a 4096 channel pulse height analyzer are used for accumulation of spectra from the data on magnetic tape. Display and software routines aid in the analysis of the spectral data. The system has been used to study environmental radiochemical analysis problems.

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INTRODUCTION

Gamma-ray spectrometric methods currently being used for measuring radionuclides in environmental materials utilize high resolution Ge(Li) detector systems and two-parameter NaI(Tl) detector systems.⁽¹⁻⁸⁾ The latter are especially useful for measurement of low levels of radionuclides which decay with the emission of coincident gamma-rays. The arrangement of a typical detector system consisting of two opposed

NaI(Tl) detectors with anticoincidence shielding is shown in Figure 1.

The natural and fallout radioactivity in soil, silt and other environmental samples interferes with the normal Ge(Li) and two-parameter NaI(Tl)-NaI(Tl) coincidence methods for detection of trace man-made radioactivity. Increased analysis sensitivity and selectivity is needed for control, monitoring, surveillance and ecological research on radioactivity in the environment. The measurement of isotopic ratios such as $^{58}\text{Co}/^{60}\text{Co}$ and $^{134}\text{Cs}/^{137}\text{Cs}$ is useful in differentiating between nuclear explosion produced and nuclear reactor produced radioactivity. (9,10)

This paper describes a multi-parameter NaI(Tl)-Ge(Li) coincidence gamma-ray spectrometry system assembled for the non-destructive analysis of trace levels of ^{58}Co , ^{134}Cs and other nuclear power industry effluent indicators in environmental samples. The system consists of high efficiency NaI(Tl) and Ge(Li) detectors, nuclear pulse-height analysis electronics and a laboratory computer for data storage and analysis. A diagram of the system is shown in Figure 2. Different detector arrangements have been evaluated for the analysis of environmental samples.

DETECTOR SYSTEMS

Two detector systems have been studied. One of the detector systems consisted of a 120 cm³ Ge(Li) detector⁽⁴⁾ opposed with a 28 cm diameter by 15.2 cm thick NaI(Tl) detector.⁽⁵⁾

Figure 1. Two-Parameter NaI(Tl) Gamma-ray Spectrometer Detector System with Anti-Coincidence Shielding

Figure 2. Diagram of Multidetector Coincidence Gamma-Ray Spectrometer

The relative efficiency (1.33 MeV at 25 cm) of the Ge(Li) detector was 26% with a resolution of 2.1 keV (FWHM) for 1.332 MeV gamma-rays. The NaI(Tl) detector had low background "Lucalox" photomultiplier tubes (RCA 4521).

The other detector system consisted of a pair of separately mounted, opposed Ge(Li) detectors surrounded by a large vertically split, annular NaI(Tl) detector.⁽¹¹⁾ The Ge(Li) detectors had relative efficiencies (1.33 MeV at 25 cm) of 12.4% and 13.1%. The resolution of the detectors was 2.6 and 2.4 keV (FWHM) for 1.33 MeV gamma-rays. The NaI(Tl) annular detector is 28 cm diameter by 23 cm long with a 10 cm diameter hole. The annulus is divided lengthwise into two optically isolated sections both contained in the same stainless steel can. Low-background "Lucalox" photomultiplier tubes (RCA 4521) were used. A diagram of the detector system is shown in reference 11.

DATA ACQUISITION

Large data arrays result from multi-detector NaI(Tl)-Ge(Li) or Ge(Li)-Ge(Li) analysis if full advantage is to be taken of the Ge(Li) detector resolution. Previous studies of coincidence spectrometry with Ge(Li) detectors were limited by the available data acquisition and storage systems.⁽¹¹⁻¹³⁾ The data acquisition and analysis system outlined on Figure 3 was assembled for coincidence spectrometry studies. The system allows the accumulation of event descriptions (ADC addresses) on magnetic tape at low count rates typical of environmental samples. After acquisition, selected data may be read back into the pulse height analyzer memory at maximum tape reading speed.

Figure 3. Four-Parameter Data Acquisition and Analysis System

The system consists of seven basic units:

- (1) Four 8192 channel ADC's.
- (2) An ADC multiplexer.
- (3) A hardwired 4096 channel, 24 bits per channel, pulse height analyzer with isometric and contour display.
- (4) Industry compatible magnetic tape drive and control.
- (5) A general purpose 16K words, 16 bits per word computer
- (6) Interfacing to interconnect the above.
- (7) Computer peripherals.

List mode data storage on magnetic tape is used for coincidence spectrometry data acquisition. The ADC multiplexer accepts data from up to four ADC's. The multiplexer then generates sequential 16-bit words corresponding to data from each ADC. Thirteen bits (8192 channels) are used for data from each ADC. Two bits are used to identify the ADC being stored and one bit indicates whether ADC rejection of a conversion occurred (as opposed to address zero due to no input). Data can be acquired in several modes including: (1) a four-input coincidence experiment, (2) a three-input coincidence experiment and one independent ADC, (3) two, dual-input coincidence experiments and (4) various other combinations of coincidence and independent experiments. Provision for rejection of non-coincident (singles) events is provided for one coincident experiment. This reduces data storage requirements when only coincidence events are of interest.

Either hardware or software list-mode operation is possible. The multichannel analyzer memory is used for buffer storage of the data in hardware list-mode. Data accumulation

is interrupted and the data recorded on magnetic tape when the 4096-word buffer is filled. The computer is completely available for other uses during hardware list-mode data acquisition. This feature is especially valuable when low-level samples are counted, since week-long counts may be routinely taken and the computer is only required for short intervals for analysis of the acquired data.

In software list-mode the multichannel analyzer memory is used for accumulation of preselected portions of any of the ADC outputs as a function of any other ADC coincident events while the computer memory is used for buffer storage of all the raw data. When filled, the 4096-word computer memory buffer is written to magnetic tape. The pulse-height analyzer and the data acquisition are controlled from the computer teletype in software list-mode. A total of 6.4×10^6 words can be stored on a reel of magnetic tape. The computer software may be stored on the front end of the magnetic tape.

Experiments are identified by a four-decimal digit tag word which is entered on the magnetic tape from either the tape controller or the teletype. Twenty-four additional bits can be written onto the magnetic tape from the teletype for each experiment.

DATA ANALYSIS

When data is accumulated under software list-mode, examination of preselected portions of the data immediately after accumulation is possible using the pulse height analyzer display.

Detailed analysis of the data may require the construction of additional portions or slices of the multidimensional spectrum from the events stored on the magnetic tape. Selected data stored on the magnetic tape can be read into the pulse height analyzer or the computer memory at high rate under software control.

Data to be examined can be selected from a single ADC as a function of specified coincident addresses or ranges of addresses in the other ADC or ADC's. Several such selected data regions may be displayed simultaneously by use of the 4K pulse height analyzer memory. 64x64 arrays can also be selected and displayed. Thus the pulse height analyzer can be used to sequentially display regions of interest for examination. Spectra constructed in core memory from the new data can also then be written to magnetic tape for subsequent rapid recall. An x-y plotter is used to make hard copy records of spectra displayed by the pulse height analyzer. Printed listings of the data can also be generated.

Software routines also provide for manipulation of the data in core memory. The centers of peaks within selected regions may be determined and the corresponding energies computed. An integration command can be used to compute peak areas. Data regions can be normalized and added to or subtracted from other regions. Bad data points may be modified via the teletype keyboard.

Single parameter spectra or single parameter slices from multiparameter spectra may be made available for search via sophisticated peak location and peak area estimation routines such as GASPAN. Such routines coupled with nuclide identification

routines can greatly reduce the man-hours required for spectral interpretation. (11,14,15)

GAMMA-RAY COINCIDENCE SPECTROMETRY

Detection of trace gamma radioactive nuclides in the presence of natural and major fallout radioactive nuclides by gamma-ray spectrometry is limited by several factors. First the radioactivity levels must be detectable above the detector system background. High-efficiency detectors are used to maximize sensitivity. Various shielding, anticoincidence and coincidence techniques have been used to reduce detector background levels. (1,2,5,6,16) The trace radionuclide photopeak or photopeaks of interest must also be resolved from the photopeaks and Compton due to other sample constituents. The use of Ge(Li) detectors has in many cases supplied the required photopeak resolution. However, Compton interference and background can still be a problem in the analysis of radionuclides at trace levels. Greater detection sensitivity is obtained by two-parameter NaI(Tl)-NaI(Tl) gamma-ray spectrometry for many radionuclides which decay with coincident gamma-ray emission. Figure 4 shows the location of the primary coincident photopeaks for the nuclides ^{58}Co , ^{60}Co and ^{134}Cs . The photopeaks resulting from ^{226}Ra , ^{232}Th and their daughters are shown to illustrate typical interference problems in natural samples. Compton as well as photopeak interference from their natural radioactivities limits the detection sensitivity; as does ^{40}K interference.

Improved resolution is possible by use of Ge(Li)-NaI(Tl) two-parameter gamma-ray spectrometry. This is illustrated in Figure 5 using the same nuclides as before. The relatively high Compton associated with the NaI(Tl) spectra can interfere with the detection of the coincident photopeaks with this detector arrangement.

Use of Ge(Li)-Ge(Li) coincidence methods is illustrated in Figure 6. This method gives excellent resolution, low backgrounds and low Compton. However, the detection efficiencies are very low. Thus very long counting times may be required. This greatly restricts the numbers and kinds of samples which can be analyzed.

Another coincidence system uses three detectors for measurement of radionuclides which decay by positron emission (such as ^{58}Co). Two NaI(Tl) scintillators detect the β^+ annihilation radiation while a Ge(Li) detector measures gamma-rays coincident with the β^+ event.

The megachannel coincidence analyzer described earlier is being used to investigate these and other coincident systems for use in environmental sample analysis. Figure 7 shows three NaI(Tl) spectra obtained by analysis of the data accumulated in a single count of a ^{60}Co source using the 120 cm³ Ge(Li)-28 cm diameter NaI(Tl) detector system described above. The spectra shown are those in coincidence with 0-2048 keV, 1329-1336 keV and 1170-1176 keV events in the Ge(Li) detector. Any other Ge(Li) or NaI(Tl) coincident conditions can be rapidly examined without recounting the sample by use of the spectral analysis software.

Figure 4. Two-Parameter Energy Regions for ^{58}Co , ^{60}Co , ^{134}Cs , ^{226}Ra and ^{232}Th Showing Photopeak Resolution Characteristic of a Pair of NaI(Tl) Detectors.

Figure 5. Two-Parameter Energy Regions for ^{58}Co , ^{60}Co , ^{134}Cs , ^{226}Ra and ^{232}Th Showing Photopeak Resolution Characteristic of a Ge(Li)-NaI(Tl) Detector System

Figure 6. Two-Parameter Energy Regions for ^{58}Co , ^{60}Co , ^{134}Cs , ^{226}Ra and ^{232}Th Showing Photopeak Resolution Characteristic of a Pair of Ge(Li) Detection.

Figure 7. Sodium Iodide Spectra of ^{60}Co , 3 Ge(Li) Coincidence Windows

The detection of ^{134}Cs in environmental samples is limited by the ^{226}Ra and daughter activities present in most environmental samples. The most sensitive method routinely used for ^{134}Cs measurement is NaI(Tl)-NaI(Tl) coincidence gamma-ray spectrometry. The ^{226}Ra interference is illustrated in Figure 4. The reduction in ^{226}Ra interference which can be obtained by use of NaI(Tl)-Ge(Li) coincidence gamma-ray spectrometry is shown in Figures 8 and 9. Figure 8 shows the NaI(Tl) spectra obtained coincident with any Ge(Li) event below 2048 keV. Figure 9 shows NaI(Tl) spectra obtained in coincidence with 603 to 607 keV events in the Ge(Li) detector. The ^{134}Cs spectrum is greatly enhanced relative to the ^{226}Ra spectrum, due to the 605 keV ^{134}Cs gamma-ray.

An evaluation of different counting modes for two-detector systems for the nuclides ^{58}Co , ^{60}Co and ^{134}Cs is presented in Tables I and II. Tables III and IV list counting efficiency and background values for selected coincidence photo-peak regions and Tables V and VI list the interference contributions from the natural radionuclides ^{40}K , ^{226}Ra + daughters and ^{232}Th + daughters for these regions. In all cases the data was obtained for 2.5-cm thick by 7.6-cm samples containing sand mixed with Scotch-Cast resin, into which each nuclide had been uniformly dispersed. From these tables it is easily apparent that where interference from activity due to natural radionuclides is serious, considerable reduction in this interference can be achieved by proper selection of detectors and coincidence requirements. A very important advantage of the

Figure 8. Sodium Iodide Spectra of ^{134}Cs and ^{226}Ra , Ge(Li) Diode Coincidence Window 0-2048 KeV

Figure 9, Sodium Iodide Spectra of ^{134}Cs and ^{226}Ra , Ge(Li) Diode Coincidence Window 603-607 KeV

TABLES I, II, III, IV, V AND VI

(In process of being completed)

system described herein is that selection of a coincidence mode need not be made prior to counting of the sample. Since all events from a given sample count are stored on magnetic tape, one can simply search the tape afterwards for combinations of events meeting the desired coincidence and energy requirements.

CONCLUSIONS

The coincidence analyzer system when coupled with dual Ge(Li), dual NaI(Tl) detectors permits simultaneous measurement in many coincidence modes. These measurements would have to be obtained in several individual counting experiments with more conventional spectrometer systems. The capability for simultaneous measurements with relatively high-efficiency NaI(Tl) detectors and with less efficient but much higher resolution Ge(Li) detectors has obvious advantages, especially for cases where it is not known beforehand what nuclides and activity levels to expect. In cases involving short-lived nuclides, it may be imperative to gather as much information as possible before activities of interest have decayed away. In cases involving nuclide interferences, only by use of NaI(Tl)-Ge(Li) or Ge(Li)-Ge(Li) coincidence methods might the nuclides of interest be measurable. In still other cases

the high resolution of the Ge(Li) detectors may be required for positive identification of the nuclides in a sample.

Choice of detector arrangements must necessarily be dependent upon the type and size of sample, the nuclides being measured, their abundances and the decay schemes involved. Trade-offs are inevitable and a considerable amount of forethought, possibly coupled with some experimentation, is required to achieve the most satisfactory measurements. For instance, the detection sensitivity for a given coincidence gamma-ray may be limited due to interference arising from higher energy gamma-rays which back-scatter from one detector to another, giving rise to two simultaneous pulses. One might minimize such events by suitable geometric arrangement of the detectors, but this usually results in reduced detection efficiency. At reduced efficiency, longer counting periods are required for low-level measurements, and thus fewer samples can be counted.

Spectrometer systems such as the one described also have application to activation analysis measurements of environmental and other samples. Nuclides which decay by coincident gamma-ray emission can often be identified in the presence of a large matrix of other activation products by use of these systems.

At the higher activity levels frequently encountered in such measurements, use of Ge(Li)-NaI(Tl) and Ge(Li)-Ge(Li) coincidence techniques permits greater selectivity than NaI(Tl)-NaI(Tl) coincidence methods, and the penalty of reduced counting efficiency is not always of great concern. An example of an application of this type is the determination of positron emitters induced by high-energy neutron bombardment of geological or meteoritic materials.

Although the large capital investment required for these systems precludes their use for routine analysis of environmental samples by some laboratories, these systems are very valuable for environmental radionuclide research studies. The nuclide detection sensitivities obtained with the coincidence spectrometry system is greater than that required for routine radiation protection measurements. Use of such systems will allow the tracing of selected effluents through the ecosystem at levels much below radiation protection standards and will permit the identification of potential problems before they reach significant levels.

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