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Lawrence Radiation Laboratory
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LIVERMORE

LOW-LEVEL COUNTING WITH SOLID-STATE $\text{Ge}(\text{Li})$ DETECTORS

Paul L. Phelps

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LOW-LEVEL COUNTING WITH SOLID-STATE Ge(Li) DETECTORS

Paul L. Phelps

Bio-Medical Division, Lawrence Radiation Laboratory,
University of California, Livermore, California 94550

The superior spectral resolution of solid-state germanium lithium-drifted (Ge(Li)) detectors as compared to that of sodium iodide (NaI(Tl)) scintillators has no doubt been responsible for their growing application to biomedical problems (1-7). The superior resolution of the Ge(Li) detector has provided a simple and accurate means of determining the energy of gamma rays associated with many of the radionuclides of interest to the biologist, who in particular has benefitted from this new analytical tool. He no longer is faced with the difficult interpretation of NaI(Tl) spectra or the tedious and often expensive radiochemical procedures that were required to be performed on samples containing complex mixtures of radionuclides.

To illustrate the superior resolution characteristics of the Ge(Li) detector, a tissue sample containing ^{65}Zn , ^{59}Fe , and ^{60}Co was analyzed first by using a NaI(Tl) scintillator and then with a Ge(Li) detector. This is shown in Fig. 1. It is obvious that the closely spaced photopeaks could not be resolved by the NaI(Tl) scintillator; but because of the superior resolution of the Ge(Li) detector, all three photopeaks were easily identified. Another case which vividly shows the usefulness of the solid-state detector

to biological problems is illustrated in Fig. 2. An experiment (8) involving the transfer of individual radionuclides from pasture grass contaminated with fission products to cows milk was easily performed.

However, one shortcoming of the solid-state Ge(Li) detector has been its lack of sensitivity, primarily due to the relatively small sizes that have been available, as compared to volumes possible with NaI(Tl) scintillators. This is particularly true for radionuclide analysis of large samples often encountered in the biomedical field. There are, however, several options open to the user of Ge(Li) detector systems for maximizing their utility for low-level counting. Spectrometers that have been developed range from the simple Ge(Li) detector configuration shown in Fig. 3 to very sophisticated systems (2, 8-9) as described in Figs. 4 and 5. The amount of complexity is, of course, dictated by the specific application and economical considerations.

In general, low-level counting as treated in this paper will imply the detection of radiation in a sample having a total activity less than 10^{-12} curies (1 pCi) or approximately 2.22 disintegrations per minute, or not more than 100 pCi for a given radionuclide contained in a complex mixture. This is to be recognized as a strictly arbitrary definition and, for example, will depend upon the sample size and the sample-detector geometrical configuration; i. e., a point source compared to a 200-cm³ sample with the same activity as the point source, relative to the surface area and depth of the Ge(Li) detector. Low-level counting systems (2, 9-10) have been developed that are capable of measuring specific activities of less than 0.05 pCi per gram with an error of $\pm 20\%$ for large 200-cm samples, and as little as a total of 1 pCi for a 2.5-cm diameter source (2, 11). Furthermore, low-level counting implies a counting system designed to detect within a reasonable

statistical limit of error, the minimum amount of radiation possible for a given application.

Basic Ge(Li) Detector

Various types of Ge(Li) detectors have been fabricated and their theory of operation described in detail (12-16). Since their basic characteristics and mode of operation are similar, a brief review can best be illustrated by referring to the "planar" type of detector shown in Fig. 6. The planar Ge(Li) detector consists of either a rectangular or a circular cross section of germanium. Lithium is initially diffused into one of the surfaces to a depth of approximately 0.3-0.5 mm. This serves as the n-type junction of the detector. The active volume of the detector is then lithium-compensated by drifting and usually is between 1 and 1.5 cm deep. The remaining undrifted germanium then becomes the p-type junction which is on the order of 1 to 2 mm thick. This can now be considered as a semiconductor diode, with the major volume having almost no free carriers bounded by a positive electrode containing no free positive holes and a negative electrode containing no free electrons. If a reverse bias voltage is applied to the two electrodes (+ H. V. connected to the n surface), there will be essentially no movement of electrical charge during a period when negligible photon interaction is taking place in the active volume. In practice, the current under these conditions is usually on the order of 10^{-10} amperes. This quiescent current must be low if very small signals are to be detected from charge produced by photons.

A comparison to a gaseous ionization chamber can probably best explain the action that takes place in the solid-state detector. If a photon enters the sensitive region of the solid-state detector (compares to the

gaseous media), free charges represented by hole-electron pairs are produced, which migrate to the electrodes (compares to ionization of the gas). The flow of charge into an external circuit then can be measured and related to the photon energy that initially produced the charge. A big difference in solid-state detectors as compared to other devices is the efficiency in which the photon energy is converted into electrical charge. In germanium a hole-electron pair is produced for each 2.9 eV of energy absorbed from the photon. In comparison it takes 30 eV per ion pair in a gaseous counter. Scintillators made from NaI(Tl) require 300-500 eV to produce an electron at the photocathode surface of the photomultiplier tube. The low conversion energy required per hole-electron pair for germanium as compared to the other materials is responsible for its superior resolution.

The photon can interact in the germanium in three different ways. What is desired is the total absorption of energy from the photon within the active volume of the detector. This occurs with the photoelectric effect, whereby nearly all the energy (minus a small amount associated with the electron-binding energy) is absorbed by an electron associated with a germanium atom. The energetic electron moves through the crystalline structure producing hole-electron pairs which are swept out by the high voltage, producing a current in an external circuit. The photoelectric effect is in competition with another process, called the Compton interaction. In this process only partial transfer of energy takes place between the photon and electron. After the Compton interaction, the degraded photon may further interact with more electrons and be totally absorbed in the germanium. However, if total absorption does not take place, the hole-electrons produced will represent some unknown fraction of the initial photon energy and

consequently have no meaningful relationship to the original photon energy. This effect is detrimental to low-level radiation counting since it causes a build-up or continuum that interferes with discrete photopeaks. A third interaction is pair-production but is of a rather small consequence for the energies of interest and will not be considered here.

Low-Level Counting

In gamma-ray spectroscopy, radionuclides are determined from a spectrum made up of photopeaks. Such a spectrum is shown in Fig. 7, where the radionuclide, ^{57}Co , has been identified. The position of the photopeak relative to the abscissa defines the radionuclide by its energy of 122 kev, characteristic of ^{57}Co . The disintegration rate or intensity of the gamma rays emitted is a measure of the amount of the radionuclide contained in the sample and is related to the number of counts shown on the ordinate. The analysis for the amount of radioactivity then requires knowing the exact number of counts contained in the photopeak. Since the ^{57}Co photopeak is superimposed on the continuum from Compton and background radiation, the actual number of counts in the photopeak will be the difference between the peak number of counts and the point of intersection between the photopeak and continuum. The ratio between the number of counts in the photopeak and continuum must be as large as possible in order to accurately determine the counts contained in the photopeak. Optimum counting precision will be a function of the square of the counting rate of the sample and directly proportional to the background rate (17, 18). This begins to get critical when the photopeak is of the same order of magnitude as the continuum. The measure of the Ge(Li) spectrometer

detection sensitivity will determine this ratio and the analytical accuracy of the system. There are several parameters in the design of a Ge(Li) gamma-ray spectrometer that must be considered in attempting to achieve sufficient sensitivity for low-level counting and at the same time maintain reasonable accuracy.

Parameters that determine the sensitivity of a low-level gamma-ray spectrometer are the Ge(Li) detector resolution and photopeak efficiency, interference from background and Compton radiation, and the counting time interval. Cooper (19) has derived an equation which relates the peak detection sensitivity of Ge(Li) gamma-ray spectrometers to the various parameters just mentioned. In general, it was shown that the minimum detectable disintegration rate, D_m , can be given to a first approximation by

$$D_m = \frac{N_m}{D_{ff} \cdot f \cdot t}$$

where N_m is the minimum number of peak counts detectable after background subtraction, E_{ff} is the detector efficiency for measuring the gamma-ray of interest, f is the fraction of disintegrations which yield this gamma ray, and t is the counting interval. The effects of these various parameters on detector sensitivity can best be understood by considering not only the detector by itself, but also the conditions under which it must be operated.

It is not only the basic Ge(Li) detector characteristics that are important, but the total environment that the Ge(Li) detector is placed in. These features are schematically illustrated in Fig. 6, which shows a detector mounted in a cryostat and totally enclosed in a lead counting shield. In addition to low-level counters using simple Ge(Li) detectors, complex

techniques have been developed and the sensitivity can be further improved for certain radionuclides by the use of Compton suppression (1, 2) and gamma-gamma coincidence (9) systems. However, the discussion to follow applies to both the simple and complex systems.

Ge(Li) Detector Parameters

Characteristics of the Ge(Li) detector that influence sensitivity, as mentioned earlier, are photopeak efficiency and resolution. The photopeak efficiency, which will be defined for this discussion as the number of counts appearing in the photopeak, compared to the total number of photons for a specific energy emitted by the radioactivity contained in the sample, is a function of atomic number (Z) of germanium, sample distance to the detector, cross section of the detector relative to the sample, and the thickness of the detector. Cooper (20) has evaluated these various parameters relative to the detection sensitivity for a variety of detector types.

Absorption of gamma rays by the photoelectric effect is proportional to the fifth power of the atomic number. The gamma-ray absorption by the process of photoelectric effect, Compton, and pair-production for germanium as a function of energy is shown in Fig. 8. The thickness of the detector will determine how efficiently gamma rays are captured as the energy is increased. The influence of detector shape on the efficiency-energy dependency and source efficiency relationship has been investigated (21, 22).

Present-day Ge(Li) detectors vary in thickness between 1.0 and 1.5 cm for the planar type shown in Fig. 3 to 4-5 cm for coaxial types. In selecting detectors for low-level counting applications the maximum detector thickness obtainable may not be the optimum choice. Since photopeak efficiency requires

capturing the maximum number of gamma rays emitted from the sample, and assuming the sample is as close as possible to the detector, the cross sectional area facing the sample must be maximized. On the other hand, gamma rays from background sources will be detected isotropically and the level of continuum interference will increase in proportion to the thickness of the detector. Also, for low-energy systems, thin detectors may be a better choice since the Compton and background continuum will be reduced.

Detector cross sections vary from approximately 20 cm^2 for high quality (resolution of 2.3 keV at 1.33 MeV) planar type detectors to 16 cm^2 for acceptable types of coaxial detectors. A summary of efficiencies for various sizes and types of detectors as a function of energy, as compiled by Camp (23) is given in Fig. 9. Also, the counting efficiency (ratio of observed counts/min to disintegrations/min) over a range of gamma energies is given in Fig. 10 for a 2.5-cm diameter source placed on the vacuum cap of the Ge(Li) gamma-ray spectrometer illustrated in Fig. 3.

A decrease in sensitivity at the low end of the energy range is caused by the attenuation of the gamma-rays as they travel through the radiation entrance window (vacuum cap) of the cryostat and the dead n-layer of the Ge(Li) detector. Also, some detector mounting designs (24) use solid material clamped over the n-layer of the detector to hold it in place and will further decrease the sensitivity to low-energy gamma rays. The choice of vacuum cap material and its thickness will determine the suitability of the system for low-energy uses. A unique Ge(Li) detector configuration developed by Goulding (25) has no appreciable entrance window ($< 1 \mu\text{m}$) and is ideal for low-energy applications. Turning a planar-type detector on its

side has also been used to reduce the attenuation of low-energy gamma rays (23).

A method of increasing the efficiency for Ge(Li) spectrometers has been reported by Saunders and Maxwell (26). They succeeded in paralleling four $7 \text{ cm}^2 \times 1 \text{ cm}$ planar-type Ge(Li) detectors without degrading resolution. The 28-cm^2 surface area gamma-ray spectrometer with a resolution of 2.91 kev was found to increase the photopeak sensitivity at 1.332 Mev by a factor of 4.8 compared to a single 7 cm^2 detector. This system was used successfully by Chertok (27) for whole-body counting of small dogs which had been administered mixtures of radionuclides. This system was successfully used for 2 years before malfunctioning of a detector developed.

Detector resolution for low-level counting is not as important as photopeak efficiency. In certain instances, however, where two photopeaks are adjacent and partially merge because of insufficient spectral resolution, the only choice may be to improve resolution. This usually implies paying a premium for the detector and incorporating a cooled first-stage field effect transistor amplifier adjacent to the detector in the cryostat. Resolution is defined as the width of the photopeak at the half-maximum level (FWHM) and is usually stated for energies of ^{60}Co (1.332 Mev) and ^{57}Co (122 kev). State-of-the-art Ge(Li) detectors that are available today have resolutions of $< 2.3 \text{ kev}$ at 1.332 Mev and $< 1.4 \text{ kev}$ at 122 kev and volumes of 20 cm^3 and 50 cm^3 for planar and coaxial types of detectors, respectively.

Background and Compton Radiation

The amount of interference from background and Compton radiation will be one of the more important factors to be considered, since it will significantly

influence the minimum amount of radiation that may be detected and analyzed for specific radionuclides.

Many materials that may be considered for the detector cryostat and counter shield will undoubtedly contain small amounts of radioactivity. Natural sources of radioactivity will be ^{40}K and the daughter products of uranium and thorium, as shown in Fig. 11. In addition, these radionuclides will be present to some extent in the building materials used in construction of the counting laboratory. Also, man-made radionuclides have been found in iron (28). Another source of radiation is from cosmic showers consisting of mesons, neutrons, electrons, and photons.

Compton radiation may be particularly troublesome if proper attention is not paid to the choice of atomic number for materials used in constructing the cryostat, since high atomic number materials will produce excessive backscatter.

Proper design of low-level gamma spectrometers requires minimizing these effects. Methods of doing this will now be discussed.

Counter Shield Construction

The function of the shield that surrounds the $\text{Ge}(\text{Li})$ detector (contained in the cryostat) is to reduce to a minimum or an acceptable level the radiation that reaches the detector from external sources. There is little problem in eliminating alpha or beta particles using rather light materials, since total absorption of these particles is affected by a thin layer of material. By far the main source of interference will be from photon radiation which attenuates exponentially as it travels through matter. Thus, any material selected to reduce gamma radiation will automatically preclude alpha and beta particles.

The most common materials in use are iron and lead. Usually the thickness selected will depend upon economic factors as well as suitable laboratory space available for the counter. Most counters in use employ lead shields with a thickness of 4 inches or iron shields 6 inches thick. Nomograms (29) are available that give attenuation factors for lead and iron shielding slabs and include the effect of build-up (the scattering of degraded radiation taken into effect) and are useful in determining thicknesses for special applications.

Low-level counting requires, no matter what material is used, that the material contain as little residual radioactivity as possible and preferably none, which is hard to achieve in practice. If lead is to be the choice, small amounts of radioactivity from uranium-thorium daughter products will be present. The amount of impurity will vary between manufacturers, and final selection should depend upon an analytical assay of the radioactivity prior to purchasing the lead.

In practice it may be easier to obtain radioactivity-free iron as compared to lead. However, thicker slabs will be required for the same gamma-ray attenuation, and other factors that will be covered later must also be considered. In most cases this will require obtaining "old" iron, since modern smelting furnaces have used ^{60}Co in the brick linings and also much of the scrap iron contains trace amounts of radioactivity. Iron from battleship armour plating or gun barrels will provide good low-level shielding. Precautions in selecting the material should be exercised and a radioassay be performed on samples of the material that is being considered for use in the shield.

Two additional factors related to the counter shield that will influence the quality of the gamma spectra generated by the detector are counter shield internal volume and the atomic number (Z) of the counter shield material.

Heath (30) has demonstrated in a qualitative manner that gamma rays that are Compton-scattered from the surrounding materials in the detector shield will produce a photopeak on the Compton distribution below the photopeak energy of interest. This will interfere with the detection of radionuclides whose gamma energies fall into the same energy region as the Compton-scattered peak. The intensity and shape of the distribution of pulses attributable to scattering is inversely proportional to the size of the shield and to the Z of the shield material. The reduction in scattering from lead as opposed to iron is due to the relatively high photoelectric absorption cross section of the high Z material. The magnitude of this problem is usually less in Ge(Li) detector systems as compared to NaI(Tl) scintillators, due to the much superior resolution and smaller volume of the former.

However, a large counter shield may not be justified if the amount of radioactive impurities contained in the shielding material is more of a detriment than the benefit gained from less backscatter with a large-volume shield.

X-Ray Fluorescence Production

If a photon is absorbed in the wall of the counter shield, it will produce an x-ray characteristic of the wall material. These x-rays may then be detected by the Ge(Li) detector, resulting in spectral interference. To minimize this effect, the counter shield is lined with materials in decreasing order of atomic number. This is often referred to as a "graded shield." The spectrometer shown in Fig. 3 is housed in such a shield. A cadmium

layer of 0.062 inches was sufficient to reduce most of the 72 kev PbK x-rays. A second layer of 0.125 inches of copper in turn reduced the x-rays produced in the cadmium. This "graded" liner very effectively reduced the fluorescence radiation to a negligible amount. In an experiment performed by Cooper (11) he has demonstrated the benefits from a shield and a graded cadmium copper liner, which is illustrated by the spectra in Fig. 11. The interfering photopeaks were mostly from ^{40}K and the daughter products of uranium and thorium contained within the walls and floor of the counting laboratory.

Cryostat Considerations

A basic function of the cryostat is to maintain the Ge(Li) detector at a low temperature. This is required in order to reduce the current from thermal excitation and prevent movement of lithium ions from the depleted region (lithium is highly mobile at room temperature). Normally, liquid nitrogen is used as the coolant, maintaining the detector at 77°K. This requires a good thermal path between the liquid nitrogen and the detector, while very good thermal insulation must be maintained between the detector and cryostat walls. This is accomplished by mounting the detector at the end of a metal rod, called the "cold finger," and immersing it in liquid nitrogen. The cold finger then protrudes into a vacuum chamber maintained at low pressure (10^{-6} to 10^{-7} mm of mercury). This is partially illustrated in Figs. 4 and 6.

Low-level counting will require two main considerations in cryostat design. First, the materials used must be free from radioactivity. The author, Cooper (11) and Bradley (31), among others, have found aluminum often to be a poor choice. Bradley also has reported traces of uranium in

beryllium material used for the radiation entrance window (vacuum cap). A magnesium detector mount shown in Fig. 6 made by Bradley (31) had no detectible amounts of radioactive contamination. This originally was reported by Cooper (11), who also has reported finding high concentrations of ^{40}K and thorium and uranium daughters in molecular sieve materials used by some manufacturers to maintain a vacuum on the cryostat. Bradley (31) has reported the best results with stainless steel and electrolytic high-purity magnesium. Under any conditions, a radioassay of materials is essential before making the final choice.

Low Z materials are to be preferred in order to reduce backscatter radiation which is proportional to Z. These materials are beryllium ($Z = 4$), magnesium ($Z = 12$), and aluminum ($Z = 13$). Other materials used frequently are stainless steel ($Z \approx 26$) and copper ($Z = 29$). The ideal choice is a cryostat containing the minimum amount of material, with the lowest Z possible and no radioactivity.

Cryostats may be designed with either a beryllium or aluminum radiation entrance window (vacuum cap). Low-energy ($< 100 \text{ kev}$) requirements may be fulfilled by using a thin beryllium window. It is possible to machine a beryllium cap to a thickness of 0.01 inches. A limiting factor in a large-diameter vacuum cap will be the amount of distortion that can be tolerated, caused by the pressure differential between the vacuum on the inside of the cryostat and atmospheric pressure on the outside.

Since the sample must be as close as possible to the Ge(Li) detector, the distance between the Ge(Li) detector and its radiation entrance window

(vacuum cap) should be no more than 3-4 mm. This is usually limited by the high-voltage stand-off required between the detector and cryostat. The author has found the sample-detector configuration shown in Figs. 3 and 6, which allows placing the sample on the surface of the radiation window, to be ideal for a large variety of biological samples.

A method to increase spectral resolution has been the employment of cryogenically cooled amplifiers using field effect transistors (FET). This method requires mounting the electronics on the cold finger as shown in Fig. 6. Considerable attention must be given to the mounting of all components to prevent mechanical movement or stress on electrical connections. In particular, two major sources of noise may be (1) the "modulation noise" by the wire that connects the detector to the amplifier caused by its vibrating and changing the electrical capacity and (2) acoustical vibrations picked up by the Ge(Li) detector. One source of noise may be the bubbling from the liquid nitrogen.

Signal Processing

The superior low-noise characteristics of field-effect transistors as compared to vacuum tubes have improved the low-level counting capabilities of Ge(Li) spectrometers. A major improvement has been the use of the cooled FET preamplifier located next to the detector. Their use is recommended when maximum system resolution is an essential requirement. It is not unusual to realize a 20-30% improvement in resolution by the use of cooled FET preamplifiers. The detailed subject of amplifier (32, 33) requirements and characteristics as they pertain to various types of detectors are adequately discussed elsewhere.

Low-level counting with solid-state detectors will require long counting periods usually ranging from 500 to 3000 minutes per analysis. This will require an extremely stable electronics system. Amplifier gain shift can be minimized to a negligible level by the use of a gain stabilization system (32) using a very stable electronic pulse as a reference signal.

Results Using the Basic Ge(Li)-Type Spectrometer

Samples were analyzed using three different detector and sample configurations to illustrate their applicability to low-level counting. All three Ge(Li) spectrometers were designed for low-level counting of biological and environmental samples (over the energy range of 60 kev to 2 Mev). All three employed planar-type detectors with cooled FET preamplifiers. Their physical configuration is illustrated in Fig. 6.

Blood Serum. A sample of blood serum containing ^{57}Co and contained in a 1.3-cm diameter glass counting vial was counted on the Ge(Li) spectrometer as shown in Fig. 3. The Ge(Li) detector was of the planar type and had dimensions of 6 cm $\times$ 3 cm by 1.2 cm thick. Its active volume was calculated to be 16.9 cm³. Detector-sample cross sectional ratio was 9 to 1, and the distance from the center of the sample to the detector surface was 10 mm, with the counting vial setting on the beryllium vacuum cap. After a counting time of 750 minutes the spectrum shown in Fig. 7 was produced. The photopeak identifying the radionuclide, ^{57}Co , is shown as a very prominent peak at approximately 122 kev. The base of the peak was at approximately 70 counts, while the peak channel had 750 counts. Total amount of ^{57}Co in the sample was determined to be 15.4 pCi $\pm$ 5 percent. Characteristics for the detector are given in Fig. 3.

Rabbit Thyroid. A rabbit thyroid containing the radionuclide ^{131}I in trace amounts was analyzed on a Ge(Li) spectrometer similar to the one shown in Fig. 3. The detector was a 1.2 cm thick planar-type with a circular cross sectional area of 9.1 cm^2 . After placing the thyroid gland in a counting vial, it was placed on the beryllium vacuum cap (0.01 inches thick). Detector-sample cross sectional area ratio was 3.25 to 1, and the sample center to detector surface distance was approximately 7 mm. Counting was performed for 912 minutes. A partial spectrum is shown in Fig. 12. The photopeak at approximately 364 kev represents the ^{131}I contained in the thyroid gland. The amount of activity was determined to be $12.8 \text{ pCi} \pm 10 \text{ percent}$, corresponding to 3.4 pCi per gram of thyroid tissue.

Air Filter Sample. An air filter sample was obtained that exhibited radioactivity from several fission product radionuclides. This sample was chosen since it represented a "bad" detector-to-sample cross sectional area geometry. The sample was 8.5 cm in diameter and 3 mm thick, while the detector diameter was only 3.5 cm or approximately one-sixth of the sample surface area. After 900 minutes of counting, the spectrum shown in Fig. 13 was produced and four photopeaks representing ^{132}Te , ^{131}I , ^{133}Xe , and ^{103}Ru were identified. Quantitation of the activity for each radionuclide was performed and is summarized in Table 1. The lowest activity detected was $8.16 \text{ pCi} \pm 25 \text{ percent}$ for ^{132}Te . Detector volume was 10.5 cm^3 , and resolutions were 2.63 kev and 1.65 kev for 1.332 Mev and 122 kev, respectively.

Soil Sample. A soil sample was obtained for analysis of radionuclides. The sample was prepared by placing it in an aluminum can (3.25 inches in diameter by 1.75 inches in height). Again this was a demonstration of "bad" detector-to-sample cross sectional area geometry. Approximate volume of

the aluminum can was 200 cm^3 , and the weight of the soil was 287 grams. Counting for 924 minutes was performed with a 3.5-cm diameter Ge(Li) spectrometer. The radionuclide ^{137}Cs was quantitated. Since the gamma-ray energy is approximately 662 kev, this represented a more difficult case compared to the previous examples, because of lower Ge(Li) detector photopeak efficiency. The difference in efficiency amounts to approximately a factor of 20 between the energies of ^{137}Cs and ^{57}Co . The amount of ^{137}Cs was determined to be $36 \text{ pCi} \pm 10.3 \text{ pCi}$. The resultant spectrum is shown in Fig. 14.

Anticoincidence Shielded Spectrometers

As mentioned earlier, the preferred gamma-ray interaction in the Ge(Li) detector is one in which the total energy of a gamma ray is absorbed by an electron of a germanium atom. The electrical charge generated as the electron moves through the crystalline structure represents the actual energy of the absorbed gamma ray. However, total absorption or photoelectric interaction competes with the Compton effect. Referring to Fig. 8, it can be seen that at an energy of 150 kev the probability of the photoelectric interaction relative to Compton absorption is 1:1, and at 1 Mev the probability is 1:100 in favor of the Compton interaction. A Compton event may deposit only part of its energy in the detector, and this appears as a continuum below the original photon energy. In time, the Compton scattering produces a continuum of increasing amplitude with a maximum at the low-energy end of the spectrum. This continuum reduces the ratio of photopeak to background, making it difficult to detect weak photopeaks as well as reducing the accuracy of determining the actual number of counts within the photopeak.

To reduce interference from Compton scattering, a large annulus made from NaI(Tl) or plastic may be used to surround the Ge(Li) detector. The theory of operation can best be understood by referring to Fig. 15. If a Compton event occurs in the Ge(Li) detector and only partial energy transfer from the incident photon takes place, the degraded photon may exit the detector. If the escape angle is such that the photon enters the anticoincidence shield, the photon will be free to interact in the shield and be absorbed. The absorption of the photon will produce a pulse of light which is then converted to an electrical current by the photomultiplier tubes. This electrical signal from the shield is then used to route the Ge(Li) detector signal to a part of the memory of the pulse-height analyzer, where all events common to both the Ge(Li) detector and anticoincidence shield will be stored. On the other hand, if total absorption of the gamma ray takes place in the Ge(Li) detector, the signal will be routed to a different part of the memory, where only those events representing total absorption will be stored. Ge(Li) spectrometer systems (1, 2) making use of this principle have been reported. A system by Phelps et al. (2) is schematically shown in Fig. 4.

For certain radionuclides, anticoincidence shielded Ge(Li) spectrometers (1, 2, 7, 9, 10, 34-38) may significantly improve the activity that can be detected in biological and environmental samples. Phelps et al. (2) have demonstrated the detection of 100 pCi of ^{137}Cs contained in 200 cm^3 of water in the presence of $0.5\text{ }\mu\text{Ci}$ of ^{60}Co which represents an extreme case of Compton interference to the ^{137}Cs photopeak. The resultant spectrum is shown in Fig. 16. Another example by Phelps et al. (2) is illustrated in Fig. 17, which demonstrates clearly the improvement for low-level counting by use of anticoincidence shielding. A sample of 200 cm^3 of water containing 10 pCi of ^{137}Cs was

analyzed by a small 6.5 cm^2 anticoincidence shielded Ge(Li) detector and a larger 18 cm^2 shielded detector. The photopeak for ^{137}Cs , representing 0.05 pCi/cm^3 was easily resolved. The Ge(Li) spectrometer with the 18 cm^2 detector was also used to quantitate ^{137}Cs contained in soil samples. The results are given in Table 2.

An exceptionally efficient and versatile anticoincidence shielded Ge(Li) spectrometer has been reported by Cooper (10) and is shown schematically in Fig. 5. A unique feature is the employment of two large (70 cm^3 and 40 cm^3) Ge(Li) detectors in an opposed geometry, completely surrounded by an anticoincidence shield. These Ge(Li) detectors may be operated also in coincidence. The high efficiency provided by the two large Ge(Li) detectors and its low background as a result of the anticoincidence shielding has yielded extremely good sensitivity. Cooper (11) has demonstrated the ability of the spectrometer to measure 1 pCi (2.2 dpm) of ^{137}Cs in the form of a 1 cm diameter source in 1333 minutes with a total error of only 10% in the presence of the natural background. Also a 0.5-inch thick by 2-inch diameter calcium sulfate disc source of 2 dpm of ^{137}Cs was reported to be easily detected with a 20% error in the presence of 1000 times more ^{60}Co in a counting interval of only 1000 minutes.

A single Ge(Li) detector employing anticoincidence shielding developed by Cooper (39) was used to quantitate the radionuclides contained in muscle tissue taken from a tuna fish caught near the Bikini Atoll in the Pacific. The resultant spectra are shown in Fig. 18. Coincidence and anticoincidence events were recorded separately. The advantage of this can be seen by referring to the ^{60}Co photopeak. Since ^{60}Co decays with cascading transitions, recording the anticoincidence spectrum only would be interpreted as if it were

Compton radiation and thus be greatly attenuated. However, recording the coincidence spectrum overcomes this; and as a result, there is no loss in sensitivity. Cooper (39) has shown a vast improvement in detecting all the radionuclides indicated. The ^{54}Mn and ^{65}Zn peak-to-background ratios were increased by almost fifty-fold, but more significantly they were not even seen in the normal spectrum.

Conclusions

Solid-state Ge(Li) detectors can be employed successfully for low-level gamma-ray analysis of biological and other types of samples. A variety of Ge(Li) spectrometer systems is available to the investigator, and the particular type will depend upon the application. Commercially available Ge(Li) spectrometers without any anticoincidence shielding will suffice for many applications. However, for the maximum sensitivity, anticoincidence shielded Compton-suppressed Ge(Li) spectrometers will be required, especially in the presence of large amounts of high-energy gamma-ray emitters.

The desired feature of a low-level Ge(Li) spectrometer will be high photopeak sensitivity with the minimum interference from background and Compton radiation. This can partially be achieved by maximizing the cross sectional area presented to the radiation from the sample. Future work should be directed toward improving the techniques for paralleling Ge(Li) detectors to achieve greater surface areas and producing very large (7-8 cm diameter) surface-area Ge(Li) detectors. This, coupled with Compton suppression methods, will greatly increase the sensitivity so that larger samples may be easily handled and reduced counting times realized.

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TABLE 1

ANALYSIS OF RADIONUCLIDES CONTAINED IN AN AIR-FILTER

SAMPLE, 8.5 cm IN DIAMETER × 3 mm THICK,

BY USE OF A Ge(Li) SPECTROMETER, 3.5 cm IN DIAMETER

<u>Radionuclide</u>	<u>Photopeak Energy (kev)</u>	<u>Radioactivity (pCi)</u>
^{132}Te	228	$8.16 \pm 25\%$
^{131}I	364	$36.9 \pm 8\%$
^{133}Xe	81	$30.1 \pm 11\%$
^{103}Ru	497	$37.1 \pm 9\%$

TABLE 2

ANALYSIS FOR ^{137}Cs IN SOIL BY THE USE OF AN
ANTICOINCIDENCE SHIELDED Ge(Li) SPECTROMETER

<u>Weight (gram)</u>	<u>Total Activity (pCi)</u>	<u>Specific Activity (pCi/g)</u>
315	34.9 ± 5.8	0.111 ± 0.018
329	30.1 ± 5.4	0.091 ± 0.017
335	36.0 ± 5.4	0.079 ± 0.017
302	28.5 ± 7.1	0.094 ± 0.024
295	37.7 ± 7.3	0.128 ± 0.025

RELATIVE ACTIVITY

ND-161, 2048
Ge (Li), 12 cm³
PLANAR, 1.1 cm THICKNESS
NaI (T_L), 3" x 3"

COUNTING TIME: 460 MIN.
GAIN: 1 keV/CHANNEL

LYOPHILIZED, 1520cc
 GEOMETRY: 200 cc CAN
 COUNTER: 13cc Ge (LID)

COUNTING TIME: 2031 MIN.
 GAIN: 1 keV/CHANNEL

1024 keV

2048 keV

0 keV

1023 keV

1024 keV

2048 keV

Peaks labeled: BG+I131, 92, Mo99, 140, Ba140, 131, 230, 238, 239, 283, 304, 328, 363, La140, 486, 536, 538, 633, 637, 664, 720, 738, 845, 921, 932, 1400, 1480, 1588, 1600.

A

B

C

ANTICOINCIDENCE SPECTROMETER SYSTEM

EFFICIENCY FOR A 2.5 cm DIA. x 0.1 cm SOURCE
LOCATED 4 mm FROM A Ge(Li) DETECTOR OF
3.5 cm DIA. x 1.2 cm.

EFFICIENCY

Counts per Channel

Counts Per Channel

GAMMA-RAY SPECTRA OF BIKINI TUNA

FIGURE LEGENDS

- Fig. 1 A comparison between a gamma-ray spectrum generated by a Ge(Li) detector and a NaI(Tl) scintillator. Note the fine spectral detail possible with the Ge(Li) detector. Counting times are not equal.
- Fig. 2 Gamma-ray spectra generated from a Ge(Li) detector. The upper spectrum represents radionuclides detected in lyophilized pasture grass containing fallout from a presumed Chinese nuclear test. The lower spectrum is from lyophilized milk from a cow grazing the pasture grass contaminated with radionuclides shown in the upper spectrum. After Potter (8).
- Fig. 3 A simple Ge(Li) gamma-ray spectrometer suitable for low-level counting. The lead shield has been lined with cadmium and copper to reduce fluorescence x-rays. Samples are placed directly on the beryllium window for gamma-ray counting.
- Fig. 4 Anticoincidence-shielded gamma-ray spectrometer for low-level counting. Detector and cryostat details are shown in the upper left-hand corner. After Phelps (2).
- Fig. 5 A dual Ge(Li) detector anticoincidence-shielded gamma-ray spectrometer of very high sensitivity. After Cooper (10).
- Fig. 6 Total "environmental" conditions under which the Ge(Li) detector must operate are important for low-level counting. This shows a simplified schematic of the detector, cryostat, and counter shield.

- Fig. 7 Analysis of blood serum for ^{57}Co by the use of a planar-type Ge(Li) detector. The ^{57}Co photopeak represents a total of 15.4 pCi.
- Fig. 8 Absorption of gamma-rays in germanium and silicon as a function of energy for photoelectric, Compton, and pair-production interactions.
- Fig. 9 Full-energy peak efficiency for various detectors located 25.4 cm from a source. The solid angle factor is included. After Camp (23).
- Fig. 10 An efficiency curve generated with a planar Ge(Li) detector 2.5 cm diameter $\times$ 1.2 cm. The source was a mixture of radionuclides contained on a 2.5 cm diameter disc and placed directly on the vacuum cap surface of the detector shown in Fig. 3. See text for explanation of efficiency.
- Fig. 11 Photopeaks produced from natural background radiation. Note the difference with and without shielding surrounding the Ge(Li) detector. The shielding uses a "graded" liner (see text). After Cooper (11).
- Fig. 12 Analysis for ^{131}I contained in a rabbit thyroid gland. The thyroid was counted on a Ge(Li) gamma-ray spectrometer similar to that of Fig. 6. The amount of ^{131}I was determined to be 12.8 pCi $\pm$ 10 percent.
- Fig. 13 An example of "bad" sample-to-detector geometry. The spectra was generated by a Ge(Li) detector which had approximately one-sixth the area as compared to the sample surface area. The ^{132}Te photopeak represents 8.16 pCi $\pm$ 25 percent.

Fig. 14 Soil was analyzed for natural and man-made radionuclides.

The soil was contained in a 200 cm³ aluminum can (3.25 inches by 1.75 inches) and counted on a 3.5 cm. diameter Ge(Li) detector. Shown here is a spectrum from 500 to 1000 kev with the ¹³⁷Cs photopeak identified. The photopeak represents 36 pCi ± 10.3 pCi.

Fig. 15 A block diagram explaining anticoincidence shielding for

Compton suppression. The method shown here records simultaneously anticoincidence and coincidence events, with spectra so derived. After Phelps (2).

Fig. 16 Spectrum from 100 pCi of ¹³⁷Cs mixed with 0.5 µCi of ⁶⁰Co in 200 ml of water. The spectrometer was an 18-cm³ Ge(Li) detector with anticoincidence shielding. After Phelps (2).

Fig. 17 Spectra from the Ge(Li) anticoincidence shielded spectrometer type shown in Fig. 4. This is an illustration of detector efficiency at the ¹³⁷Cs energy for Ge(Li) detectors of 6.5 cm³ (left) and 18-cm³ (right) surface area. The sample, 10 pCi ¹³⁷Cs in 200 ml water was counted for 1000 minutes in each case. It was impossible to detect the ¹³⁷Cs without anticoincidence shielding. After Phelps (2).

Fig. 18 Coincidence and anticoincidence gamma-ray spectra of muscle tissue from a tuna collected in the region of the Bikini Atoll. The ⁵⁴Mn and ⁶⁵Zn were of such low levels that they could not be detected in the normal (non-Compton suppressed) spectrum. After Cooper (39).

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