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**Portable Instrumentation for
Quantitatively Measuring Radio-
active Contamination Levels and
for Monitoring the Effectiveness
of Decontamination and
Decommissioning Activities**

R. L. Brodzinski

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June 1983

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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PORTABLE INSTRUMENTATION FOR QUANTITATIVELY
MEASURING RADIOACTIVE CONTAMINATION LEVELS AND
FOR MONITORING THE EFFECTIVENESS OF DECONTAMINATION
AND DECOMMISSIONING ACTIVITIES

R. L. Brodzinski

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PORTABLE INSTRUMENTATION FOR QUANTITATIVELY MEASURING
RADIOACTIVE CONTAMINATION LEVELS AND FOR MONITORING THE
EFFECTIVENESS OF DECONTAMINATION AND DECOMMISSIONING ACTIVITIES

R. L. Brodzinski

SUMMARY

Two completely portable high-resolution germanium diode spectrometer systems are described. These detectors are capable of measuring transuranics, activation products, and fission products, including ^{90}Sr , at sensitivities below the uncontrolled release criteria. The detectors measure x-rays, gamma-rays, or bremsstrahlung radiation as required and have been calibrated for a variety of decontamination and decommissioning scenarios. A description of a new technology for the in-situ determination of ^{90}Sr is given.

INTRODUCTION

Starting with the Manhattan Project, the nuclear industry, both defense and commercial, has created large quantities of equipment, facilities, and buildings contaminated with radioactive materials. Specific examples include walls, ceilings, and floors of buildings, glove boxes, fabrication and process equipment, piping and ventilation systems, and large volumes of sediments which received process wastes either deliberately or accidentally. Modern commercial nuclear power plants are likewise responsible for producing large amounts of contaminated equipment, such as the failed Surry steam generator and the debris from the accident at Three Mile Island.

Although the total volume of contaminated waste is somewhat staggering, the amount of radioactive material involved is relatively small. Most of the contamination is attached to surfaces, even in the case of contaminated sediments where the radioactivity can be considered to be on the surface of the individual grains of sand. The primary goal of decontamination efforts associated with decommissioning activities is to remove the radioactive materials from the contaminated surfaces of the surplus facilities and equipment so that the bulk of the material may be disposed of as noncontaminated waste, or even better, reused or recycled.

In order to measure the effectiveness of decontamination efforts, a quantitative analysis of the radiocontamination is necessary, both before and after decontamination. Since it is desirable to release the decontaminated material for unrestricted use or disposal, the assay equipment must provide adequate sensitivity to measure the radioactivity at or below the release limit. In addition, the instrumentation must be capable of measuring all kinds of radiocontaminants including fission products, activation products, and transuranic materials. Finally, the survey instrumentation must be extremely versatile in order to assay the wide variety of contaminated surfaces in many environments, some of which may be extremely hostile or remote.

This report describes the development and application of portable instrumentation capable of quantitatively measuring most transuranics, activation products, and fission products, including ^{90}Sr , on almost any contaminated surface in nearly any location.

TECHNICAL APPROACH AND INSTRUMENTATION

The basic approach applies high resolution germanium diode spectroscopy to the measurement of photons emitted by the radioactivities. Nearly all of the radioisotopes of concern in decontamination activities emit some type of photon radiation, and those few that don't can be inferred from the presence or absence of accompanying nuclides that do emit photons. Most activation and fission products emit copious quantities of gamma-rays of characteristic energy which can be used to both identify and quantify particular species. Strontium-90, a common fission product, and its equilibrium daughter product, ^{90}Y , emit only beta particles, but these electrons produce significant amounts of fairly energetic bremsstrahlung photons during their deceleration in the matrix material. A few transuranic materials emit gamma-rays of sufficient energy and intensity that they can be identified in the same manner as fission and activation products. However, most transuranic activities are alpha emitters with only weak branching fractions to high energy states which de-excite by emission of gamma-rays. All transuranics emit copious quantities of x-radiation, however, which is of sufficient energy and intensity to be readily measurable with a high resolution germanium diode spectrometer designed specifically for that purpose. Because of the low energy of the abundant x-rays and the low intensity of high energy gamma-rays, the quantitative measurement of low concentrations of transuranic materials in the presence of high concentrations of fission or activation products is normally not possible. Fortunately, due to the methods of formation and the general nature of the industry, measurement of transuranic contaminants in the presence of high concentrations of fission or activation products is normally not required.

Two germanium diode spectrometers, specifically adapted to portable, multipurpose use, were acquired to provide measurement capability for both gamma-rays and x-rays. One is a large volume intrinsic germanium coaxial detector having a FWHM resolution of 1.82 keV at the 1332 keV line of ^{60}Co and a relative efficiency of 32.8%. The other is a 2000 mm² planar intrinsic germanium detector housed in an end cap with a thin beryllium window and having a resolution of 739 eV at the 122 keV line of ^{57}Co . Both crystals are mounted in 12-inch long end caps and attached to portable liquid nitrogen cryostats with nominal holding times of 16 hours. These diodes are shown in Figure 1.

Data acquisition is normally accomplished with a 4096 channel analyzer, Davidson Model 4106 A, and transferred to cassette tape for subsequent computer reduction and analysis. In semi-remote locations, power is supplied by a portable Honda Model EM 1500 gasoline generator, and in extreme conditions a totally self-contained battery powered system is mounted on a backpack frame. The backpack electronics package with

detector and remote control switch is shown in Figure 2, and the system is shown in operation in Figure 3.

Data reduction is normally done with the aid of a PDP 11-44 computer either following "reading" of the data stored on cassette tape or, if immediate results are required and a telephone line is available, by direct connection to the laboratory based computer. The procedures for identifying radioisotopes on the basis of their characteristic gamma-ray energies and for determining their concentrations by comparison of their characteristic line intensities to those in reference spectra are well established. Similarly, transuranic elements, but not individual isotopes, can be identified and quantified on the basis of their characteristic K or L shell x-ray emission lines. The only difficulty in these procedures is obtaining suitable calibration data. The calibration data must be acquired under nearly identical measurement conditions and with radio-contaminants of known concentration. In many instances a calibration "curve" or family of curves is used to interpolate calibration factors on the basis of photopeak intensity, matrix density, or attenuating materials.

The case for quantitative determination of ^{90}Sr is not as simple as for the gamma-ray emitting radionuclides, however, and a more detailed discussion of this new technology will be given here. Since the bremsstrahlung radiation spectra generated by ^{90}Sr represents a continuum of photon energies with no distinguishing monoenergetic photopeaks, such as observed from most other fission product radionuclides, the relationship between photon intensity and ^{90}Sr concentration is somewhat harder to determine. Furthermore, many other fission products likely to be present in ^{90}Sr contaminated environments also emit energetic beta particles which contribute to the total bremsstrahlung spectrum and which must be accounted for prior to assignment of bremsstrahlung radiation to ^{90}Sr . Hence, it was decided that the most accurate approach to data reduction would be to "strip out" each identifiable component from an unknown spectrum until only ^{90}Sr bremsstrahlung radiation remained. This method assumes the compilation of a library of photon spectra from each pure radionuclide, for each detector to be used, and for each matrix material to be investigated. It further assumes that ^{90}Sr is the only pure beta emitter present with sufficiently energetic beta particles to contribute to the residual bremsstrahlung spectrum at a significant level. This latter assumption is valid in virtually all practical situations. The former assumption, though valid, would require a tremendous amount of baseline data acquisition, all of which may not be necessary in practical situations. An empirical approach to a common field situation was chosen as a demonstration of the true capability of the technique.

Since it is virtually impossible to construct a calibration facility representing an infinite volume of homogeneously contaminated undisturbed sediments, and since it is impractical to artificially generate large volumes of contaminated sediments in order to characterize those already inadvertently contaminated zones, a different approach to the development of a calibration facility was taken. A cursory survey

of the Hanford site yielded a suitably located pristine area which had never been tilled or disturbed in any other way by man. This area was designated the Natural Activities Calibration Facility and was appropriately marked to prevent inadvertent intrusion. The surface vegetation and soil were sampled using a statistically acceptable geometrical pattern, and the samples were carefully analyzed in the laboratory for all measurable radioactivities. The only radionuclides present were primordial ^{40}K , uranium and thorium and their progeny, and a trace of ^{137}Cs attributed to fallout. The concentrations of the primordial activities indicated the site was spatially homogeneous to an accuracy of $\pm 8.3\%$. A six-inch schedule 40 steel well casing was inserted near the center of the site by means of vacuum coring, and the core material was collected in 6-inch intervals. Samples of each 6-inch layer were also analyzed in the laboratory, and the primordial radionuclide concentrations were consistent within $\pm 5.7\%$. The known concentrations of homogeneously distributed radionuclides were used to generate an empirical efficiency curve as a function of photon energy for our system. This efficiency curve is shown in Figure 4. A photograph of one of the spectrometers being calibrated at the NACF is shown in Figure 5.

The next step was to create a reference library of pure radionuclide spectra, including man-made activities, without contaminating the site. Sealed sources of known intensity were fabricated for each radionuclide of interest including the primordial activities ^{226}Ra and ^{228}Th . The thorium source was inserted into the sediments at the NACF, and the net spectrum due to the standard source was measured. The source depth was then repositioned, and the procedure was repeated until the net spectrum due to the thorium standard had precisely the same shape as the primordial ^{228}Th component of the background. The comparison of spectral shape was made on the basis of relative intensity of all photopeaks in the spectrum. With the source in the same position, the radium standard also demonstrated the primordial spectral shape. Hence, this point source location was used to generate "replicate" library reference spectra of man-made radioactivities which represent the expected response to a uniformly contaminated zone of undisturbed sediments. Note that these secondary calibration spectra are not necessary, nor used, to determine the concentration of gamma-ray emitting radioisotopes since these calibrations are directly available from the energy-efficiency curve determined on the basis of the primordial activity concentrations. These library spectra are necessary, however, in order to "strip" an unknown field spectrum down to the ^{90}Sr bremsstrahlung component. A standard ^{90}Sr bremsstrahlung spectrum was also obtained from the point source location in order to compare the intensity and hence determine concentration of an unknown.

Although a library of reference spectra was now available, it was far from a simple task to strip out the various components and determine the residual ^{90}Sr bremsstrahlung activity. First, the background had to be stripped from each reference spectra so that all data represented photo events from a pure radioisotopic contamination. This led to the first data handling problem. If the background is correctly subtracted

from a spectrum, there should be no net activity at any energy above the highest energy photopeak produced by the radioisotope -- but, there should be a statistical fluctuation above and below zero counts. Our first modification to the computer data handling software allows the entry of negative events in a gamma-ray spectrum, a maneuver not normally encountered during gamma-ray spectral accumulation. However, if the computer were allowed to truncate all negative numbers to zero, then future subtraction of reference spectra would result in an over-subtraction from an unknown spectrum. The next data handling modification was to realign a field spectrum as a function of photon energy to allow for incorrect calibration in the field or for zero or gain drift during the course of data acquisition. The difficulty in establishing and maintaining gain stability in a field environment which may involve extreme heat or cold and portable generator or battery supplied electric power is a nontrivial problem. Simplistically, the gain shift modification divides the field spectrum into individual counts and redistributes them into new locations based on the relative slope of a calibration line drawn through two of the prominent peaks in the field spectrum and the true calibration line drawn through the precise energy of the same photopeaks. The total number of counts is not changed in this process, but the spectrum is redrawn with all photopeaks appearing at precisely the correct energy. This data manipulation technique does not correct for loss of resolution due to field effects or gain shifts during the count, but this minor drawback has proven to be only a cosmetic effect and does not affect the data reduction accuracy.

The complete procedure for deconvoluting an unknown field spectrum starts with obtaining a background spectrum which can ordinarily be done in a similar but uncontaminated zone. After gain adjustment, the background spectrum is multiplied by the ratio of live counting times and subtracted from the unknown spectrum leaving only man-made radioisotope contributions. Each radioisotope contributing to this spectra is identified on the basis of its characteristic full energy photopeaks, and the corresponding reference spectra are called from the computer library, normalized on the basis of photopeak intensity, and stripped out of the unknown spectrum. The full energy photopeak intensities in the unknown spectra are, of course, also used to determine the concentration of each radioisotope in the matrix. The residual unknown spectrum, after stripping, represents only ^{90}Sr bremsstrahlung radiation and is compared to the referenced ^{90}Sr spectrum on the basis of intensity to determine concentration. Four different energy regions are normally compared in order to ensure that the residual spectrum has the correct ^{90}Sr bremsstrahlung shape, and that each energy region yields the same value for the ^{90}Sr concentration. In the event that no ^{90}Sr is present in the matrix, the residual spectrum should appear as a straight line with only a statistical fluctuation of data above and below zero. Finally, a smoothing function has been incorporated in the software which can be applied to any spectrum for cosmetic purposes. This smoothing function neither creates nor destroys counts, but merely re-assigns neighboring channel intensities in order to remove the sharp spikes engendered by many subtractions and subtraction of peaks of the same intensity but different resolution.

The only purpose of this smoothing function is to render the final product more palatable to the untrained eye which is unaccustomed to seeing negative numbers in gamma-ray spectra.

Another commonly encountered field situation is a contaminated surface wherein the radioactivity does not penetrate deep into the matrix material. Calibration of the germanium diodes for this circumstance was accomplished by using point sources of x-ray and gamma-ray emitting standards distributed across an "infinite" plane divided into a large number of concentric rings having equal area. The germanium diodes were placed at the center of these concentric rings an appropriate distance away from the plane, and the point sources were counted for an equal period of time in each ring. The net spectra were then summed for all rings out to "infinity" (the point which no longer added a significant contribution to the total spectrum), and the efficiency curve as a function of photon energy was determined based on the point source activity divided by the area of one ring. In practice, it was found that the exact spectral shape and intensity could be reproduced in a single measurement at a given distance from the center of the circles (a distance of 4.24 m with the diode located 1 m above the surface), and this approach was used to gather the large number of spectra necessary. As in the contaminated sediments exercise, a ^{90}Sr reference spectrum was also acquired for surficial contamination. The efficiency curve for determination of surficial concentrations of gamma-ray emitting radionuclides is shown in Figure 6.

In a field situation where it is not known whether the radiocontamination is surficial or whether it has penetrated into the matrix, the data provide the answer. The spectral shape is influenced by the amount of photon scattering encountered and, hence, the amount of matrix material intimately associated with the activity. The peak-to-Compton ratio will be higher for a gamma-ray emitting radioisotope located on a surface than the corresponding ratio for the same isotope located within a matrix. Similarly, the peak-to-Compton ratio will be even lower yet if the activity has been covered over with uncontaminated material. Appropriate calibrations provide quantitative measures of these peak-to-Compton ratios and, hence, identify the location of a radiocontamination.

RESULTS AND DISCUSSION

The application of the x-ray detection system to the determination of plutonium and americium on soil surfaces and pond bottoms has been published elsewhere⁽¹⁾ and will not be repeated here. Detection limits for these experiments were 2.8 pCi cm⁻² for ^{241}Am and 33 pCi cm⁻² for ^{239}Pu at soil depths up to several millimeters. Americium-241 can be measured at soil depths of several centimeters at a sensitivity of 4.6 pCi cm⁻². A similar application of the TRU x-ray detection system to the in situ analysis of contaminated laboratories, process hoods, and buildings⁽²⁾ found detection limits similar to those given above.

A pictorial example of the application of gamma-ray spectrometry to the characterization of a surface contaminated with ^{137}Cs and ^{90}Sr is

demonstrated in the following. Figure 7 shows the actual spectrum as acquired. A major ^{137}Cs component is apparent to even the most casual observer. Just as apparent to the experienced observer is a major ^{90}Sr component, which will be demonstrated in the following sequence. First, a background spectrum obtained in a similar, but uncontaminated, environment is subtracted from the unknown spectrum after both have been "gain shifted" to the proper energies. The net spectrum is shown in Figure 8, and the ^{137}Cs concentration is deduced from the intensity of the 661.649 keV photopeak. Some minor variations in resolution and gain between the unknown and background spectra are apparent at the major primordial photopeak areas. Figure 9 shows a smoothed version of Figure 8. The smoothed version is not used for continuing data reduction -- it is only a demonstration of an intermediate step. Also, since no significant data exists above 1600 keV, that portion of the spectrum will be omitted in the following figures. The net ^{137}Cs reference spectrum is shown in Figure 10, with a smoothed version shown in Figure 11. This reference spectrum is normalized to the same intensity as the unknown spectrum on the basis of the 661.649 keV photopeak and is subtracted out leaving only ^{90}Sr bremsstrahlung radiation as shown in Figures 12 and 13. The net ^{90}Sr reference library spectrum is shown in Figures 14 and 15. Comparison of the bremsstrahlung intensity in the unknown and reference spectra yields the ^{90}Sr concentration directly. The detection limit for ^{90}Sr in normal operations with this bremsstrahlung measurement technique is less than the release limit which is 60 pCi cm^{-2} . The detection limit for ^{137}Cs , a representative gamma-ray emitting fission product, is 6.6 pCi cm^{-2} which is nearly an order of magnitude below the release limit of 60 pCi cm^{-2} .

The Natural Activities Calibration Facility is being maintained in order that other field assay and well logging instrumentation may be simply and inexpensively calibrated. Several surface assay detectors and well logging germanium diode gamma-ray spectrometers have already been calibrated at the site, and the same technique of data handling and reduction can be used to measure ^{90}Sr concentrations in subterranean environments as well as to determine the depth of ^{90}Sr activities which have been covered over with uncontaminated materials.

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FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

FIGURE 6

FIGURE 7

FIGURE 8

FIGURE 9

FIGURE 10

FIGURE 11

FIGURE 12

FIGURE 13

FIGURE 14

FIGURE 15

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