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RADIATION DOSIMETRY THROUGH SPECTRAL DEFINITION

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ABSTRACT

We have developed a fieldable instrumentation system for determining from measured flux spectra, both the neutron and gamma ray dose rate distributions associated with radioactive sources. This system includes the sensors, the computer-based data acquisition and analysis hardware, and the requisite software for unfolding the sensor response functions to obtain the flux spectra, and for folding the resultant flux spectra with appropriate flux spectrum-to-dose conversion factors. We use bismuth germanate scintillators that have experimentally measured and analytically interpolated response functions to determine the gamma ray flux spectra, and a suite of neutron sensors, based on proton recoil and ^3He capture to determine the neutron flux spectra. In addition, gamma ray peak identification is done using HPGe sensors. We describe the equipment and procedures and present some recent results.

INTRODUCTION

The increased concern about radiation exposures in general, and the specific expectation of changes in the quality factors for nuclear radiations and the effects of these changes on reported exposures, have prompted us to approach the problem of radiation dosimetry in a somewhat unconventional fashion. Instead of using only standard integrating dosimeters that are assumed to have, or have been certified to have, an energy response function that mimics one of the flux-spectrum-to-dose conversion curves, we measure the flux spectra of the radiations and perform operations on these spectra. We either fold these with one of the several accepted flux-spectrum-to-dose conversion factors (Fig. 1) to arrive at the dose rate values or use these spectra to correct the response of integrating dosimeters. Obviously, to obtain the spectra requires the use of high sensitivity sensors that have been well characterized, and microprocessor-based data acquisition and analysis systems that have sufficient capability to acquire the pulse height distributions, unfold the sensor response functions, and perform the convolutions with the flux-spectrum-to-dose conversion curves.

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GAMMA-RAY SYSTEMS

For our purposes, the sensors chosen for the gamma-ray spectrum determination are bismuth germanate (BGO) scintillators and high-purity germanium (HPGe) diodes. The latter are used to best characterize the radioactive materials involved using the high resolution capability of HPGe. The time required to acquire these high resolution spectra is often unacceptably long in many applications because the efficiency of a single, large, HPGe detector with good resolution is typically only 40% of that of a NaI(Tl) detector 7.6 cm in diameter by 7.6 cm long.

To improve the efficiency, we built a gamma-ray spectrometer system that consists of an array of three 40% high-purity germanium detectors and a computer to digitally sum the three spectra without significant loss of resolution (Fig. 2). The three detectors in the array have relative efficiencies of 43.0, 44.5, and 40.0% and resolutions of 1.77, 1.76, and 1.71 keV (FWHM) at 1332 keV, respectively. Each detector has its own all-attitude dewar so that it can be used separately. In the array, the three detectors are held in position by a styrofoam and aluminum frame of low mass to minimize scattering and absorption.

The computer program that combines the distributions calculates a highly accurate calibration for each distribution. The gains for the distributions can be quite different. First the program locates several strong peaks with accurately known energies using an automatic peak-search routine; then it fits the peaks. Manual interaction is possible. The program calculates a least squares fit to determine the calibration. The calibration curve can be any function; however, tests showed that a straight line was adequate with our analog-to-digital converters. If the spectra do not contain enough strong peaks, the program uses recent calibrations from another source.

The program then compresses the two distributions with the highest gains to match the distribution with the lowest gain. The compression of each distribution requires summing fractions of the number of counts in each channel to generate a new distribution. The calibrations are required to calculate these fractions.

To calculate the final composite distribution, the program performs a channel-by-channel sum of the two compressed distributions and the distribution with the lowest gain. The composite distribution is then further analyzed.

The high efficiency of the HPGe array promotes the use of high-resolution gamma-ray systems for these applications. However, for dosimetry purposes, such high resolution is not required and actually presents logistical difficulties because the HPGe detectors must be cooled, many channels must be used to acquire the pulse-height distributions, and the response functions are complicated. We avoid these problems by using BGO scintillators for dosimetric purposes.

The BGO system, which is packaged for easy shipment, consists of eight 7.62-cm-diam. by 7.62-cm-long scintillators and a LeCroy 3500 data acquisition system (Figs. 3 and 4). Bismuth germanate is a good choice for the scintillators because of its high efficiency and large peak-to-total ratio that simplifies the analysis. We calibrated and characterized the system from 0.12 to 8.29 MeV with gamma rays from radioactive sources and reactions in Van de Graaff targets. By simultaneously fitting several calibration spectra with a function containing 17 parameters, we were able to construct response functions. We then use these functions to obtain the gamma-ray flux spectra at multiple points in the radiation field. The results of our procedure agree with calculated values to within less than 10%. Details of these calculations are given in Refs. 2 and 3. This method of parameterizing the bismuth-germanate response functions also can be used to calculate the performance of proposed systems that contain bismuth-germanate scintillators.

One application of the BGO system was the determination of flux distributions and dose rates from containers of uranium and plutonium. Figure 5 shows results from a 550-g shell of depleted uranium containing 0.2% ^{235}U . Prominent peaks from the $^{234\text{m}}\text{Pa}$ daughter occur at 767, 1001, and 1800 keV. Much of the gamma-ray output from uranium is bremsstrahlung, which is responsible for the continuum out to the 2.3 MeV endpoint. Figure 6 shows results from a 455-g shell of plutonium containing 2.65% ^{240}Pu . Complexes of gamma-ray lines produce peaks at 414, 640, and 760 keV. The dose rates, which are more accurate than those obtained with simple energy-dependent dosimeters, are useful when dose rates must be known accurately because of safety or legal considerations.

A second application was the determination of the flux distribution and dose rate from a replica of Little Boy, the device exploded over Hiroshima. The replica was operated at low power as a reactor, at the Los Alamos Critical Assemblies Facility. To minimize scatter the device was supported outside on a stand so that the core was 4.0 m from the ground when operating. Figure 7 shows some results at 0.75 m from the center and 90° from the vertical cylindrical axis. The distributions are dominated by the 7.631-7.645 MeV doublet from neutron capture in the massive steel case. Because data obtained from the Hiroshima and Nagasaki survivors provide the basis for many of our radiation safety guidelines, it is important to establish the radiation exposure of these survivors of the Hiroshima explosion. Experimentally determined flux and dose rates, such as these, provide checks of the calculations at low power and increase the confidence in the calculations for the explosion.

NEUTRON SYSTEMS

The response of neutron dosimeters is also dependent on energy. Measurement of neutron spectra is therefore necessary to accurately determine dose rates directly or to determine correction factors for neutron doses measured with integrating neutron dosimeters and dose-rate measuring instruments. For neutron energies from 0.6 to 20 MeV, proton-recoil spectrometry with liquid scintillators is the normally used technique. For lower energies, gas-filled proton-recoil proportional counters are used. The ^3He ionization chamber operating in the energy range from 20 keV to 2 MeV offers better energy resolution (14 keV for low-energy neutrons, 30 keV at 1 MeV) than proton-recoil counters over most of its range and the pulse height distribution is usually easier to unfold. We have characterized these detectors using both Van de Graaff and electron linear accelerator generated neutrons and have applied all three of these techniques to the measurement of neutron spectra.

Neutron spectra below 1 MeV

An example of our work in neutron spectrometry below 1 MeV is given by the measurement of the leakage spectrum from the Little Boy replica.⁴ A high-resolution gridded ^3He ionization chamber of the type built by Cuttler and Shalev³ was used to obtain spectra in the energy range from 20 to 300 keV. Subsequently, information from 150 to 900 keV was obtained using a proton-recoil gas proportional counter that we are developing.

The gridded ^3He ionization chamber has a sensitive volume 5 cm in diameter by 15 cm long filled with 5 atm of ^3He , 1 atm of argon, and a small amount of methane. The stainless steel counter tube is surrounded by a thermal-neutron shield consisting of 2 mm of pressed boron nitride sandwiched between two 0.5 mm sheets of cadmium.

Details of the procedures used to calibrate and characterize this detector are found in Ref. 5. Briefly, the spectrometer was calibrated with a set of response functions for monoenergetic neutrons of energies from 100 to 1200 keV. The peak-efficiency curve was extrapolated below 100 keV using the inverse-velocity dependence of the $^3\text{He}(n,p)$ cross section. Pulse shape discrimination is used to eliminate some of the effects of competing reactions, such as ^3He recoil, from the pulse height distributions. The data acquisition and analysis system used for the neutron spectrometry is microcomputer-based of the type shown in Fig. 4.

In Fig. 8, the Little Boy neutron spectrum from 0 to 1 MeV is shown. Also shown in the figure is the ENDF/B-V total neutron cross-section for iron. All of the indicated peaks in the spectrum correspond to prominent minima in the iron cross section. In particular, approximately 15% of the neutrons above the epithermal peak are to be found in the 24-keV iron window, which is often used to produce a nearly monoenergetic 24-keV neutron beam at reactor facilities. The results are consistent with the thickness of iron that the neutrons penetrate.

We have been developing a line of proton-recoil proportional counters to measure neutron spectra below 1 MeV. These counters, patterned after a suggestion of Verbinski and Giovannini⁶ are cylindrical chambers 7.5 cm in diameter by 30 cm long, with hemispherical ends (Fig. 9). Anode, guard-tube, and insulator dimensions are proportioned according to Benjamin et al.⁷ to minimize end effects. The counter was filled to a pressure of 200 mm of mercury with 80% H_2 , 18.4% xenon, 1.5% methane, and 0.1% ^3He . The energy scale and resolution of the spectrometer system were determined from the position and width of the ^3He peak when the detector was irradiated with thermal neutrons.

A set of response functions was measured, using monoenergetic neutrons at the Los Alamos 3.75-MeV Van de Graaff. A program was written to strip these response functions, starting with the highest energy response, from an experimental pulse-height distribution to produce a spectrum with resolution characteristic of the detector.

In Fig. 10, a pulse-height distribution from the Little Boy replica has been fit by a sum of response functions. The solid line is the experimental pulse-height distribution. The areas between curves are the contributions to the distribution of each of the monoenergetic responses. Counting statistics were too poor above 960 keV, and the number of events above this energy was too small to materially affect results at lower energies.

In Fig. 11, the results from the proton-recoil counter are compared with those from the ^3He spectrometer. The ^3He spectrometer results have been binned into the same energy intervals as the proton-recoil spectra for comparison. The ^3He and proton-recoil counter measurements compared well in the region of energy overlap with the results of liquid scintillator spectrometry and with integral measurements with Bonner spheres and other devices.

For neutron sources with strengths comparable to ambient levels, we have found the above detectors insufficiently sensitive. We are now studying the possibility of using helium gas scintillators for some of our measurements. One of these detectors, shown in Fig. 12, filled with 200 PSIG of either ^3He or ^4He , is 30 times as efficient as either of the gas counter tubes, making possible in hours what now takes days to measure. These detectors are relatively insensitive to gamma radiation and have extremely short pulse lengths, of the order of ten nanoseconds. However, they lack the high energy resolution of the ^3He ionization chamber and there is no pulse-shape difference between ^3He capture and ^3He recoil pulses. It is felt, however, that in many cases where ultimate energy resolution is not required, the helium gas scintillator with its higher data acquisition rate may be a preferred instrument.

Neutron Spectra Above 1 MeV

We have used the proton-recoil liquid scintillator, NE 213, to measure neutron spectra in the range of 1 to 10 MeV. Because this detector is also sensitive to gamma-rays, pulse shape discrimination is required to eliminate pulses from the data that are produced by gamma-ray interactions. This pulse-shape discrimination depends on the longer decay tails on pulses produced by neutron interactions. We use different ADC channels to separately integrate the full pulse and the pulse tail. The ratio of the full pulse area to the area of the pulse tail then produces a parameter which is used to discriminate neutron and gamma ray interactions in the scintillator.

The pulse height data are then unfolded using the NE 213 response functions measured by Verbinski et al⁸ and the "least structure" unfolding method.⁹ In this method the square of the difference between the measured data and the refolded solution is minimized along with the square of the second derivative of the solution. This is done to damp extraneous, unbounded solutions which can be introduced into the result by small errors in the data or by truncation or rounding errors.

The details of our methodology are to be found in reference 10. The data acquisition and analysis system used in the NE 213-based neutron spectrometry program is microcomputer based, of the type shown in Fig. 4.

In one of the applications, the neutron flux spectrum from the Little Boy replica was measured using a 5 cm-diam. by 5 cm-high cylindrical NE 213 liquid scintillator detector. Because of this detector's sensitivity to gamma rays, we surrounded it with 1.3 cm of lead to reduce the event rate due to gamma-ray interactions in the detector. In addition, we used the pulse-shape discrimination (PSD) method to identify and later eliminate pulses produced by gamma-ray interactions.

The results from two measurements made on the Little Boy replica appear in Figs. 13 and 14. These results are presented without compensation for the effect of the lead used to reduce the gamma-ray event rate in the detector. Both measurements were made with the leakage flux incident on the curved surface of the cylindrical detector reproducing the geometry used by Verbinski in measuring the detector response functions.

To obtain the dose or dose-rate spectra from these neutron flux spectra, we simply fold the flux spectra with the neutron flux-spectrum-to-dose conversion factors.

Because we have been concerned that the historical Verbinski response functions for NE 213 scintillator cells may not be accurate for our specific detector, we have measured them for the 5 cm diameter x 5 cm long cell used in the Little Boy replica work.

We used the Oak Ridge Electron Linear Accelerator and time-of-flight techniques to obtain the pulse height distributions as a function of incident neutron energy. The neutron energy range spanned was from 300 keV to 25 MeV. This range was divided into approximately 70 bins of 4096 channels of pulse height data each. We are constructing response function matrices for these detectors and will use them in the future to improve both the accuracy and energy resolution in our NE 213 work. Some of our preliminary results are shown in Fig. 15 which is a comparison of neutron spectra measured by time-of-flight and pulse height unfolding. The time of flight spectrum has been corrected for neutron detector efficiency. The unfolded spectrum is from our preliminary response functions. The very good agreement is an indication that the method we used for the Little Boy replica was accurate.

CONCLUSIONS

Nuclear radiation dose rates cannot be measured directly: at best they can be calculated as the convolution of measured neutron and gamma ray spectra and the radiation flux-spectrum-to-dose conversion factors. The typical approach, however, is to use health physics survey instruments that provide readings of dose rates. These instruments have spectrum sensitive response functions, requiring a knowledge of the spectrum for interpretation of the readings. We have adopted the method of measuring the neutron and gamma ray spectra and determining the dose rates by the convolution method.

The radiation spectra, themselves, however, are not directly measurable either, but the response of suitable energy sensitive detectors immersed in these radiation fields is directly measurable. These responses are typically obtained as pulse height distributions. If the response functions are measured then the radiation flux spectra can be obtained by unfolding.

Once the flux spectrum is known for each of the radiation types of interest, the appropriate weighting with the energy-dependent flux-spectrum-to-dose conversion factors will yield the dose rates resulting from these emanations.

We have carefully followed a course of characterizing both gamma-ray and neutron sensors and developing the data acquisition and analysis systems to obtain pulse height distributions, to unfold using precisely known response functions that are used to yield the radiation spectra and then to fold these spectra with appropriate flux-spectrum-to-dose conversion factors to finally obtain the dose rate spectra. Having archival radiation spectrum information offers the possibility of recalculating exposure doses in the event there are changes in the flux-spectrum-to-dose conversion factors as a result of reevaluations of quality factors.

We have used our transportable stand-alone system to make numerous dose rate measurements, examples of which are given in this paper. In addition, we have used the spectrometry capability to provide correction or weighting factors for conventional, integrating type health physics instruments.

REFERENCES

1. P. J. Dimbylow and T. M. Francis, "A Calculation of the Photon Depth-Dose Distributions in the ICRU Sphere for a Broad Parallel Beam, A Point Source, and an Isotropic Field," National Radiological Protection Board report R92, Harwell, England (1979).
2. C. E. Moss, E. J. Dowdy, A. E. Evans, M. E. Hamm, M. C. Lucas, and E. R. Shunk, *Nucl. Instr. and Meth.* 219 (1984) 558.
3. C. E. Moss, E. W. Tisinger, and M. E. Hamm, *Nucl. Instr. and Meth.* 221 (1984) 378.
4. A. E. Evans, "Little Boy Neutron Spectrum Below 1 MeV," Proceedings of the Seventeenth Mid-year Topical Symposium of the Health Physics Society, 8.29-8.36 (1984).
5. J. M. Cuttler, S. Shalev, and Y. Dagan, "A High-Resolution Fast-Neutron Spectrometer," *Trans. Am. Nucl. Soc.* 12(1):63 (1969).
6. V. Verbinski and R. Giovannini, "Proportional Counter Characteristics and Applications to Reactor Neutron Spectrometry," *Nucl. Instr. and Meth.* 114 (1974) 205-231.
7. P. W. Benjamin, C. D. Kemshall, and J. Redfearn, "A High Resolution Spherical Proportional Counter," *Nucl. Instr. and Meth.* 59 (1968) 77-85.
8. V. V. Verbinski, W. R. Burrus, T. A. Love, W. Zobel, N. W. Hill, and R. Textor, "Calibration of an Organic Scintillator for Neutron Spectrometry," *Nucl. Instr. and Meth.* 65, (1968) 8.
9. A. A. Robba and L. R. Veaser, "A Least Structure Unfolding Code," Los Alamos Scientific Laboratory report LA-5088 (November 1972).
10. A. A. Robba, "Measurement of the High-Energy Leakage Neutron Spectrum From the Little Boy Mockup," Los Alamos National Laboratory report (to be issued).

FIGURE CAPTIONS

Fig. 1.

Gamma-ray flux-spectrum-to-dose-rate curve based on the work of Dimbylow and Francis.

Fig. 2.

Three-detector array and associated electronics. As shown, the detectors can be used individually. The LeCroy 3500 is the unit on the right with the video display. The large box on the left contains a CAMAC crate and a NIM bin. A modem is under the telephone. The case on the floor contains a terminal and a PEGASUS computer with a floppy disk for communicating with other computers.

Fig. 3.

Array of eight BGO scintillators supported on tripods around a drum that contains radioactive material. The disposable grid taped to the floor facilitates positioning the detectors.

Fig. 4.

LeCroy 3500 data acquisition system and NIM bin electronics. The shipping box under the NIM bin electronics is for the LeCroy system.

Fig. 5.

Binned gamma-ray pulse-height distribution (top), flux distribution (middle), and dose-rate distribution (bottom) from a 550-g shell of depleted uranium. Each major peak is labeled with the energy in keV and the associated radioactive nuclide.

Fig. 6.

Binned gamma-ray pulse-height distribution (top), flux distribution (middle), and dose-rate distribution (bottom) from a 455-g shell of plutonium.

Fig. 7.

Binned gamma-ray pulse-height distribution (top), flux spectrum (middle), and dose-rate spectrum (bottom) at 90°, 0.75 m from the Little Boy replica. The 7.6-MeV peak from $\text{Fe}(n, \gamma)$ is the most prominent feature of the distribution.

Fig. 8.

Neutron spectrum from 0- to 1-MeV compared with the ENDF/B-V total cross section for iron.

Fig. 9.

Proton-recoil proportional counter for intrinsic radiation measurements.

Fig. 10.

Little Boy proton-recoil spectrum fit with response functions for monoenergetic neutrons.

Fig. 11.

Proton-recoil and ^3He -spectrometer results at 90°, 0.75 m.

Fig. 12.

High-pressure helium gas scintillator assembly.

Fig. 13.

Waist measurement of the neutron flux spectrum at 2 m from the core center.

Fig. 14.

Nose measurement of the neutron flux spectrum at 2 m from the core center.

Fig. 15.

A comparison of neutron spectra measured by time-of-flight and pulse height unfolding.

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Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

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Figure 8

Figure 9

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Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15