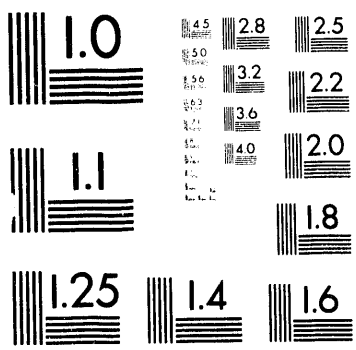

1.0
1.1
1.25
1.4
1.6
1.8
2.0
2.2
2.5
2.8
3.2
3.6
4.0
4.5
5.0
5.6
6.3

1 of 1

UCRL-JC-112823
PREPRINT

Monte Carlo Simulations of Plutonium Gamma-Ray Spectra

Zachary M. Koenig
Joseph B. Carlson
Tzu-Fang Wang
Wayne D. Ruhter

This paper was prepared for submittal to the
34th Institute of Nuclear Materials Management Annual Meeting
Scottsdale, Arizona
July 18-21, 1993

July 16, 1993

MONTE CARLO SIMULATIONS OF PLUTONIUM GAMMA-RAY SPECTRA

Zachary M. Koenig, Joseph B. Carlson, Tzu-Fang Wang, and Wayne D. Ruhter,
Lawrence Livermore National Laboratory, Livermore, California, USA*

Abstract

Monte Carlo calculations were investigated as a means of simulating the gamma-ray spectra of Pu. These simulated spectra will be used to develop and evaluate gamma-ray analysis techniques for various nondestructive measurements. Simulated spectra of calculational standards can be used for code intercomparisons, to understand systematic biases and to estimate minimum detection levels of existing and proposed nondestructive analysis instruments. The capability to simulate gamma-ray spectra from HPGe detectors could significantly reduce the costs of preparing large numbers of real reference materials. MCNP was used for the Monte Carlo transport of the photons. Results from the MCNP calculations were folded in with a detector response function for a realistic spectrum. Plutonium spectrum peaks were produced with Lorentzian shapes, for the x rays, and Gaussian distributions. The MGA[1] code determined the Pu isotopes and specific power of this calculated spectrum and compared it to a similar analysis on a measured spectrum.

Introduction

Using Monte Carlo calculations to simulate gamma-ray spectra of special nuclear materials, as compared to generating spectra from experiments, is faster, cheaper, safer, and most of all, helps reduce waste. Once the simulation is established, a calculated standard allows system-performance capabilities to be determined much faster than if an actual standard is made for every geometry and media. This simulation can supplement measurements with actual standards and give a good estimate of assay performance. In addition, the cost of running a simulation calculation is significantly less than the cost of preparing, measuring, and disposing of standards. Perform these calculations, as compared to working in a laboratory with radioactive isotopes, makes the entire endeavor inherently safer because of no radiation exposure. Finally, this capability minimizes waste because there is no laboratory waste to dispose of.

*Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48 and the DOE Office of Safeguards and Security

Overview

Our Pu gamma-ray simulation calculation consisted of three parts. First, the source term describes the gamma rays emitted from the radioactive isotopes. The computer code, GAMGEN,[2] developed at Lawrence Livermore National Laboratory calculates associated gamma-ray (from the isotopes and their daughters) intensities.

The second part is the gamma-ray transport: the source term plus the description of the source-detector geometry served as an input file for the MCNP Monte Carlo radiation transport code.[3] Results from the calculation simulate gamma-ray energy deposition in the detector volume.

The third and final part of the simulation calculation is incorporating the detector response function. This simulates a realistic gamma-ray spectrum measured by a multichannel analyzer. The detector response function includes Gaussian broadening of the peaks based on the number of events, electronic noise, and charge loss effect due to a non-ideal detector. These effects can be determined by a number of different methods.

One method includes measuring the pulse-height spectra resulting from a large number of monoenergetic sources and forming a response matrix as a function of pulse height and source energy from simplistic interpretations of the measured spectra. Another method uses a Monte Carlo simulation to model the energy deposited and charge collection resulting from the major mechanisms. A number of authors[4,5] have developed Monte Carlo approaches to study response functions of Ge detectors. Jin, *et al.*[6] developed a semiempirical model for the response function of Ge detectors based on fundamental interactions mechanisms. A third method describes the various features of a spectra by fitting a measured spectra to analytical functions. A least-squares estimate of the peak-shape parameters of these analytical forms is obtained as a function of energy. Helmer and Lee[7] review the models and background subtraction procedures of most currently used codes. The MGA program by Gunnink[1] was developed specifically to analyze the multiple peaks of the plutonium spectrum. This analysis was used for our application.

Peak shape characterization

Measured spectra from a HPGe planar detector determined the peak-shape parameters for the gamma rays. The peak-shape parameters include not only the Gaussian shape but also the tailing terms, due to a non-ideal detector and signal processing. Tailing occurs primarily on the low-energy, and occasionally on the high-energy, side of a peak. Low-energy tailing results from incomplete charge collection in the detector, and/or low-angle scattering in the sample or in the surrounding materials. Tailing on the high-energy side arises from signal distortions by the pulse electronics modules. High-energy side tailing had a minimal effect on the peak shape with proper electronics setup and count rate. The low-energy-side tailing portion of a peak can be described by one or more exponential terms. The computer code, MGA,[1] determined the low-energy-side, peak-shape parameters of the planar detector.

Calculated plutonium spectrum-radiation source term

We used an extensive database of the precise energies and branching-ratio intensities that included all of the decay chain and gamma-ray line data for all the nuclides in the four heavy-element decay series. This included the decay chain data of Browne and Firestone,[8] along with the IAEA-recommended gamma-ray energy and emission intensity values for ^{235}U, ^{239}Pu, and ^{241}Am.[9] The energies and emission intensities for ^{237}U, $^{238,239,240,241}Pu$, and ^{241}Am from Gunnink, *et al.*[10] were also incorporated. The minor differences that existed between the IAEA and Gunnink data were resolved in favor of the latter. Figure 1 shows the discrete lines and relative intensities produced from GAMGEN for the 92–104-keV region of the Pu spectrum. For our work, the mixture of nuclides included $^{238,239,240,241,242}Pu$ and ^{241}Am. The source emissions for the principal nuclides in Pu comprise in excess of 2100 lines as well as bremsstrahlung continua. It was necessary to select only the most prominent discrete x- and gamma-ray lines for transport by MCNP as discrete lines and accumulate the large number of remaining lines into a considerably smaller number of energy bins, also for transport. There were 170 discrete lines chosen for transport, and an additional 125 equally spaced binned data composed of all the remaining lines up to 3 MeV.

Radiation transport - MCNP

The geometry of the Pu source and its containment was fully modeled in three dimensions, along with the detector geometry up to the back of the detector housing. The geometry of the source and detector arrangement is shown in Figure 2. This was the same geometry also used for the measured spectra. The upper energy cutoff was 3 MeV. The MCNP volume source option, which randomly starts photons throughout the interior of the sample, was used so that the code simulated the physical transport process.

After the random distribution of events in direction and energy, the photons are attenuated through the material surrounding the source and detector. This attenuation is based on a database of attenuation coefficients for the various materials. The amount of time needed to calculate enough statistics can be reduced if only events emitted in the approximate direction of the detector and above a given energy are followed to completion. Energy-deposited tallies were performed for the detector volume. The Monte Carlo problem ran long enough to get a sample precision of a few percent in all of the significant energy bins. These calculations were run on a UNIX-based SUN workstation.

Detector response

MCNP and other general-purpose Monte Carlo codes are not well suited for simulating gamma-ray spectra with all of the attributes of a detector response function. The energy deposition tally with a simple Gaussian broadening is the extent to which MCNP comes to creating a realistic gamma-ray spectrum. It is necessary to include all of the peak-shape characteristics in the post-processing to accurately describe the peak shapes in the 94–104-keV energy region used by the MGA code.[1] This region contains peaks from all of the Pu isotopes of interest. They are also the most intense peaks above 59 keV. The more than 15 x rays and gamma rays of this region form a very tight cluster of peaks, as shown in Figure 3, from a measured Pu spectra.

X-ray peak shapes can differ significantly from those of gamma rays with the same energies because atomic transitions generally have much shorter lifetimes (i.e., with measurable Lorentzian width) than nuclear transitions in the actinide decays. It is important to recognize that the energy distributions of these intrinsic radiations is

Lorentzian in shape. The Lorentzian-broadened x-ray distribution is convoluted with the nearly Gaussian instrumental response.[11] The result of this combination is a far more complex peak shape than those produced by gamma rays. Several authors[12,13,14] have developed methods of convoluting a Lorentzian signal with a Gaussian instrumental response that include both numerical and analytical approximations. The detector response should include not only the Gaussian approximation but also the tailing terms necessary to describe a non-ideal detector. The Lorentzian distributions should first be applied to the discrete x-ray lines before the radiation transport calculation, but because the Lorentzian line widths are only of the order of 100 eV, the variation in the transport results were negligible. Figure 4 shows the calculated spectra with the appropriate peak-shape parameters included for the 94–104-keV energy region used by the MGA code.

Measured plutonium spectrum

The Pu sample used for these measurements was a PuO_2 (6% ^{240}Pu) pressed pellet encapsulated in a welded stainless-steel container. The Pu mass in the sample was about 0.45 grams. An EG&G planar HPGe detector obtained the 0–300-keV gamma-ray spectrum of Pu. These spectra were used for experimental comparisons to the calculated plutonium spectra. The same geometry from the calculation was used for the measurement. Figure 3 shows the measured spectra taken with the HPGe detector.

Comparison of computed and measured spectra

The MGA was used for both the calculated and measured spectra to determine the Pu isotopes and specific power. The fits of the 94–104-keV region of the measured and calculated spectra are shown in Figures 3 and 4, respectively. The residuals (Figure 4) in the Monte Carlo simulation show a wiggling behavior. This can be partially due to (i) the lack of statistical accuracy in the simulated gamma-ray backgrounds, and (ii) the inability of the code to simulate a realistic Compton profile. The errors of the background from the Monte Carlo simulation with our limited computation time are in the order of 10–20%. At present, we are running the same simulation with 10 times more particle tracks than the reported simulation. The results from the higher statistic run will clarify the origin of the wiggling behavior in the residuals.

The purpose of the MGA analysis is to determine the relative amounts of Pu isotopes, hence the specific power. This is used in conjunction with a measurement from a calorimeter to determine the total amount of Pu by nondestructive analysis for safeguards. Table 1 compares the calculated and measured relative Pu isotopes and specific power.

Conclusions

We have demonstrated that the complex features of a Pu gamma-ray spectra can be applied to a Monte Carlo transport calculation. The resulting gamma-ray spectra has sufficient detail to be used by the isotopic analysis code, MGA. These calculated spectra can replace experiments with an understanding of the limitations that include well-known geometries and source composition.

References

1 R. Gunnink and W.D. Ruhter, *MGA*, UCRL-LR-103220, Vols. 1-2, Lawrence Livermore National Laboratory, Livermore, CA, 1990.

2 T.B. Gosnell, *Nucl. Inst. and Meth.*, **A299**, 682, 1990.

3 J.F. Breismeister, ed., LA-7396-M, Rev.2 Manual, Los Alamos National Laboratory, Los Alamos, NM, 1986.

4 K.M. Waino and G.F. Knoll, *Nucl. Inst. and Meth.*,**44**, 213, 1966.

5 R. Chun, L.L. Gadeken, and B.C. Robertson, *Nucl. Inst. and Meth.*, **137**, 295, 1976.

6 Y. Jin, R.P. Gardner, and K. Verghese, *Nucl. Inst. and Meth.*, **A242**, 416, 1986.

7 R.G. Helmer and M.A. Lee, *Nucl. Inst. and Meth.*, **178**, 499, 1980.

8 E. Brown and R.B. Firestone, *Table of Radioactive Isotopes*, Wiley, NY, 1986.

9 Technical Report Series N. 261, IAEA, Vienna, Austria, 1986.

10 R. Gunnink, J.E. Evens, and A.L. Prindle, UCRL-52139, Lawrence Livermore National Laboratory, Livermore, CA, 1976.

11 K. Debertin and W. Pessara, *Nucl. Inst. and Meth.*, **184**, 497, 1981.

12 D. H. Wilkinson, *Nucl. Inst. and Meth.*, **95**, 259, 1971.

13 C.C. Batty, S.D. Hoath, and B.L. Roberts, *Nucl. Inst. and Meth.*, **137**, 179, 1976.

14 P.L. Lee, *Nucl. Inst. and Meth.*, **144**, 363, 1977.

Table 1. Weight percent of plutonium isotopics and specific power.

	Standard value	Measured value	Measured error	Calculated value	Calculated error
^{238}Pu	0.0111	0.0106	0.0002	0.0121	0.0025
^{239}Pu	93.7962	93.7826	0.0188	94.1443	0.50
^{240}Pu	5.9933	5.9792	0.021	5.6397	0.40
^{241}Pu	0.1994	0.2026	0.0005	0.1789	0.03
^{241}Am	0.2314	0.2215	0.0007	0.2408	0.01
Specific power	2.550	2.552	0.002	2.565	0.025

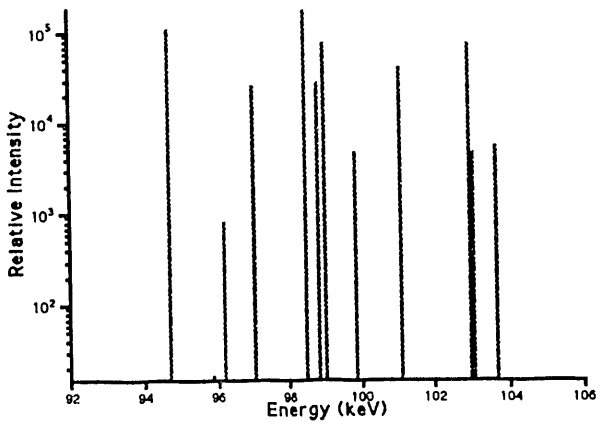


Fig. 1. Relative intensities of prominent discrete gamma-ray from plutonium generated by GAMGEN.

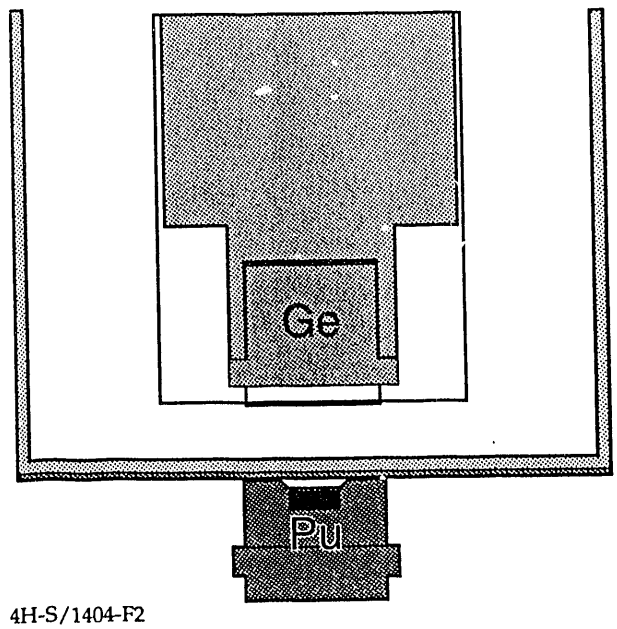
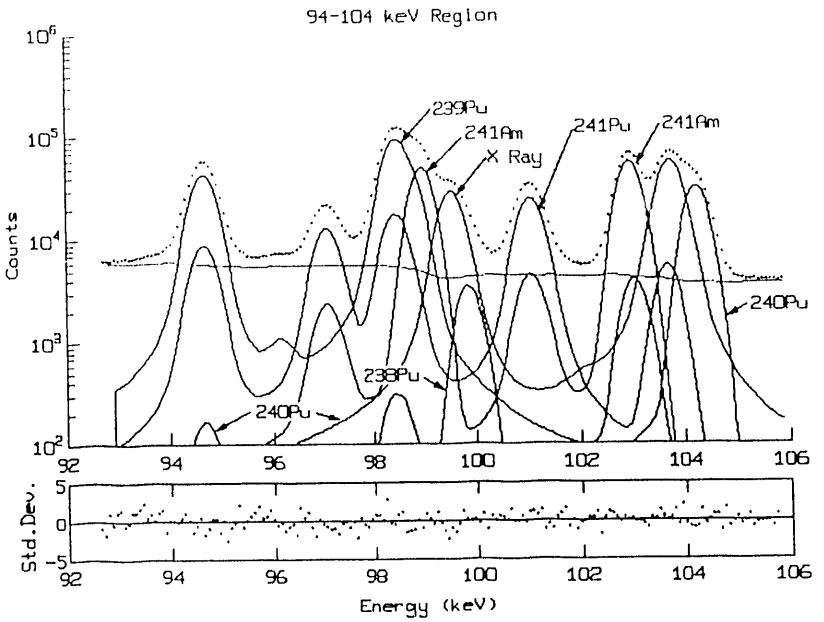
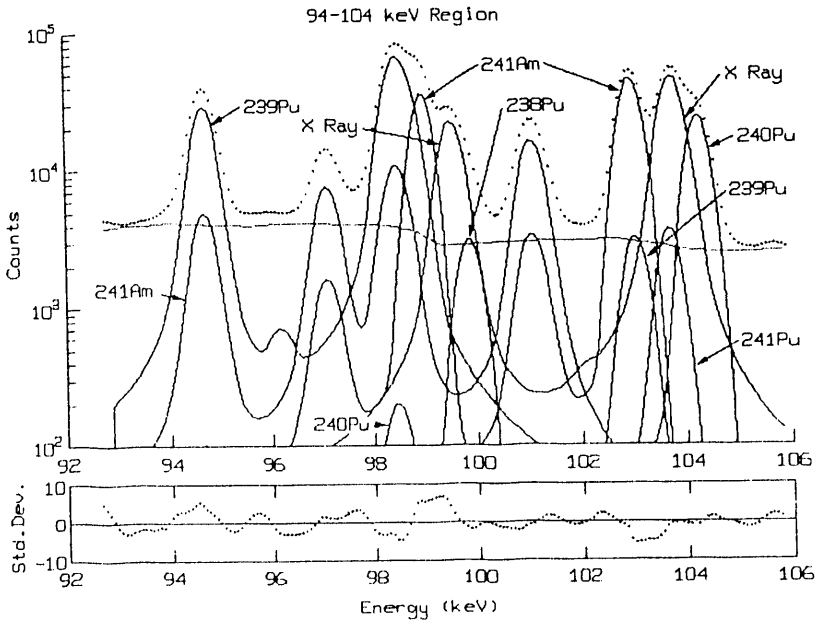


Fig. 2. Geometry description of source and detector for MCNP input.



4H-S/1404-F3

Fig. 3. The 94–104 keV energy region for the measured Pu gamma-ray spectrum.



4H-S/1404-F4

Fig. 4. The 94–104 keV energy region for the MCNP Pu gamma-ray spectrum.

DATE
FILMED
12/14/93
END