Preprint
UCRL-JC-133130

Plutonium and Uranium Isotopic Analysis: Recent Developments of the MGA++ Code Suite

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This article was submitted to
6th International Meeting on Facilities Operations - Safeguards Interface, Jackson Hole, WY, September 20-24, 1999

September 17, 1999

U.S. Department of Energy

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PLUTONIUM AND URANIUM ISOTOPIC ANALYSIS: RECENT DEVELOPMENTS OF THE MGA++ CODE SUITE

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ABSTRACT

The Lawrence Livermore National Laboratory develops sophisticated gamma-ray analysis codes for isotopic determinations of nuclear materials based on the principles of the MultiGroup Analysis (MGA). MGA methodology has been upgraded and expanded and is now comprised of a suite of codes known as MGA++. A graphical user interface has also been developed for viewing the data and the fitting procedure. The code suite provides plutonium and uranium isotopic analysis for data collected with high-purity germanium planar and/or coaxial detector systems. The most recent addition to the MGA++ code suite, MGAHI, analyzes Pu data using higher-energy gamma rays (200 keV and higher) and is particularly useful for Pu samples that are enclosed in thick-walled containers. Additionally, the code suite can perform isotopic analysis of uranium spectra collected with cadmium-zinc-telluride (CZT) detectors. We are currently developing new codes with will integrate into the MGA++ suite. These will include Pu isotopic analysis capabilities for data collected with CZT detectors, and U isotopic analysis with high-purity germanium detectors, which utilizes only higher energy gamma rays. Future development of MGA++ will include a capability for isotopic analyses on mixtures of Pu and U.

I. INTRODUCTION

MGA++ is a suite of computer codes that analyzes gamma-ray spectra and determines the relative isotopic abundances of actinides in a sample. The original MGA code was developed to determine plutonium isotopic abundances for gamma-ray data collected with germanium detectors. The MGA++ code suite includes MGA as well as several other codes for isotopic analysis of uranium and plutonium samples. The MGA++ code suite is fully modularized. The first code in the code suite an upgraded version of MGA which performs plutonium isotopic analysis using the 100 keV energy region. The second code is U235, a uranium isotopic analysis code which uses gamma rays less than 200 keV to determine uranium isotopic abundances. The third code is CZTU, a uranium isotopic analysis code which uses gamma rays less than 200 keV collected with a cadmium-zinc-telluride (CZT) detector. The fourth code in the suite is Pu600, a plutonium isotopic analysis code which uses the 200 keV – 700 keV energy region. Pu600 determines the Pu relative isotopic ratios for ^{239}Pu, ^{240}Pu, and ^{241}Am and sets a lower bound on the mass of ^{239}Pu. With the exception of CZTU, the codes analyze gamma-ray data collected with an HPGe detector.

Additionally, new codes are being developed. The fifth code in the suite is MGAHI, a plutonium isotopic analysis code which uses the 200 keV – 1 MeV energy region. This code has been completed recently and is being used at Lawrence Livermore National Laboratory. MGAHI is particularly important for situations in which the lower energy gamma rays cannot be seen. Three other codes are under development. U235HI will use higher-energy gamma rays to determine uranium isotopic content in a uranium sample. MGAUPU will use the MGA methodology to analyze mixtures of uranium and plutonium. CZTPu will analyze plutonium data collected with a cadmium-zinc-telluride detector.

Two graphical user interfaces have been developed for viewing data and the fitting procedure. The first is linked in with the MGAHI code. The second graphical user interface is a separate package and can be used with any of the

American Nuclear Society, Jackson Hole WY,September 1999
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MGA codes. This second package is called GRFXSVR (graphics server).

Several of the codes have been under development via a cooperative R&D Agreement (CRADA) between Lawrence Livermore National Laboratory and EG&G ORTEC. The codes MGA, U235, and CZTU have been developed under the CRADA. Also, CZTU has also been licensed to Canberra Industries.

II. DESCRIPTION OF THE COMPUTER CODES

The MGA methodology, including detailed descriptions of peak shapes, efficiencies, geometry considerations, and background subtraction is described in detail in Refs. 1–5. The only data provided to the codes are gamma-ray energies and branching intensities, half-lives of the isotopes, mass absorption coefficients, and the spectrum data. The basic method for determining the relative isotopic abundance is to measure the intensity of two or more peaks from gamma rays of similar energy, but arising from different isotopes. Because the gamma-ray emission probabilities and half-lives are known, the atom ratios can be calculated if relative detection efficiencies for the peaks can be estimated. For many samples the counting geometry or the counting efficiency is not reproducible, and the gamma-ray attenuation by the sample matrix or other absorbing materials is not known. When the energies are nearly equal, the efficiency and attenuation differences are small, and the ratio of the isotopic abundances can then be directly related to the peak intensities by

$$A_1/A_2 = (I_1 \lambda_2 B_2 \varepsilon_2 t_2) / (I_2 \lambda_1 B_1 \varepsilon_1 t_1), \qquad (1)$$

where A_1/A_2 = isotopic ratio, I = measured peak intensity of isotope, λ_1 = decay constant of isotope, B_1 = branching ratio of isotope one , ε = gamma-ray counting efficiency, t = gamma-ray transmission to the detector.

I_1 and I_2 must be determined with a high level of accuracy to get precise isotopic ratios.

The local intragroup relative efficiencies can be determined accurately by delineating intergroup efficiencies. The curve describing intergroup efficiencies, commonly known as the intrinsic efficiency curve,[6] is determined by fitting observed peak intensities to a function used to describe the relative efficiency with energy.

Gamma rays are fitted with a Gaussian profile and a low energy exponential tail. X-rays are fitted with a Voight profile, the shape resulting from the Lorenzian profile emitted by the x-rays and the Gaussian detector response. For each code, techniques were developed to unfold the complex peak multiplets observed in the spectra using mathematical descriptions of the peak shapes.

A. MGA

MGA analyzes gamma-ray data collected with an HPGe detector and reports the relative isotopic ratios of all isotopes of plutonium except ^{242}Pu. The value of ^{242}Pu is calculated with an algorithm or may be input by the user. The most recent version of MGA has a new method of ^{242}Pu isotopic determination.[6] MGA also calculates the isotopic abundances of ^{241}Am, ^{235}U, and ^{238}U when present along with plutonium. The code gives a U/Pu ratio, and can handle ^{243}Am-^{239}Np and ^{241}Am inhomogeneities. MGA will analyze both freshly separated and aged samples. MGA will analyze data collected with a LEPS detector. MGA can also analyze

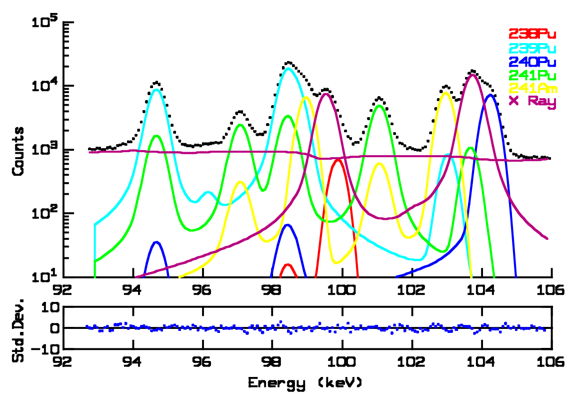

Figure 1. MGA analysis of the 100 keV energy region for a plutonium sample. The spectrum shows peaks from ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, and x-rays. This spectrum illustrates the complexity of the region and the accuracy of the MGA methodology.

MGA is one of the codes which has been under development via a cooperative R&D agreement (CRADA) between Lawrence Livermore National Laboratory and EG&G ORTEC. Additionally, the most recent version of MGA has been supplied to Euratom in Luxembourg for use in European safeguards work. Along with the MGA software which was delivered to Euratom, we developed a weeklong MGA experts class which was taught in Luxembourg in March 1999.

B. MGAHI

The MGAHI analysis is similar to that of MGA but has been improved to overcome the limitations encountered by MGA with shielded samples. MGAHI uses physical parameters to take into account both attenuation and emission of the gamma rays, and does not require a detector efficiency calibration. The gamma-ray information between 50 keV and 200 keV is not required.

MGAHI is useful when sources are heavily shielded, and in a hot, space-limited environment. With too much shielding, the 100 keV energy region may be completely attenuated. In a hot environment, the detector may see gamma rays from other nearby sources. Also, the detector may have a very high dead time. Pb shielding may be used in this situation to reduce the dead time, but this may also cut out the low energy gamma rays.

MGAHI analyzes data collected with one COAX HPGe detector. The software requires the gamma-ray information from 200 keV to 1 MeV to determine the Pu isotopic information. A detector resolution of 1.1 keV or better at 208 keV is required. The 203-keV gamma ray from ^{239}Pu must be visible. Up to 5 mm of Pb absorber can be placed in front of the detector. Detector efficiencies, absorber thickness and Pu thickness are calculated from the spectral data using known gamma-ray peaks from the decay of ^{239}Pu. During development of MGAHI, the gamma-ray attenuation of plutonium, cadmium, iron and lead has been re-parameterized using up-to-date evaluated data.

A simple Windows/NT graphical interface the shows the three main fitting regions and results, as well as a plot of the spectrum and the relative efficiency. This graphics interface is linked in with the code and shown illustrated in Fig. 2. There are four sections of Fig. 2. The top left section shows the gamma-ray energy spectrum, with the efficiency curves. The other three sections of Fig. 2 show the important regions used the MGAHI fitting. The regions used the MGAHI analysis are the 325 – 350 keV energy region, the 360 – 385 keV energy region, and the 635 – 670 keV energy region. Each of the three fitting sections shows the data, the fits to the data, and the residuals. Examination of the residuals for a fit gives an instant determination of the quality of fit.

Table 1 shows results of some calibration Pu standards from the MGAHI fitting compared to their declared values. Data were collected using a 75% COAX detector for 3 hours per measurement. Two absorbers, 5 mm stainless steel (SS) and 2 mmPb, were used. Results from destructive analysis (DA) are also tabulated.

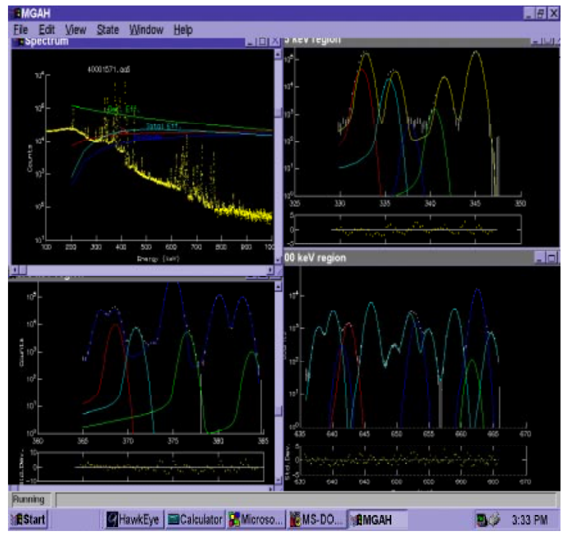

Fig. 2. The Windows/NT graphical interface of MGAHI. The upper left picture is the spectrum data. The other three pictures are the fits to the data in the three important energy regions. For each of the three regions, the residuals is plotted and shown here.

	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
PIDIE#1 + SS	0.012 ± 18%	93.79 ± 1%	6.02 ± 4%	0.19 ± 7%
PIDIE #1 + Pb	0.011 ± 15%	93.85 ± 1%	5.98 ± 3%	0.18 ± 5%
PIDIE #1 DA	0.01108	93.822	5.969	0.1975
PIDIE #3 + SS	0.042 ± 14%	84.65 ± 1%	14.34 ± 3%	0.97 ± 4%
PIDIE #3 + Pb	0.044±1 ± 3%	84.91 ± 1%	14.04 ± 3%	1.01 ± 3%
PIDIE #3 DA	0.0475	84.835	14.128	0.99

Table1. MGAHI Pu weight % results of the two Pu (PIDIE) standards. Results with lead and stainless steel (SS) absorbers and declared values (DA) are reported.

Future development of MGAHI will include extending the analysis capability down to 100 keV for situations in which the material container or shielding may be thick.

C. U235

The U235 analysis code determines the percentage of ^{235}U, ^{238}U and ^{234}U in a uranium sample from the analysis of the emitted gamma rays.[8] ^{235}U and ^{238}U sample analysis is complicated in that the gamma rays observed often come from their radioactive daughters produced by successive alpha and beta decays. In addition to gamma decay, these elements decay by internal conversion (IC), and subsequent emission of daughter product x-rays. Also, x-rays are produced by gamma rays interacting via the photoelectric effect in the material itself producing fluorescent x-rays.

The most important region in the ^{235}U analysis is the 87 – 100 keV energy region. This region, shown in Fig. 3, has three peaks due to ^{238}U, a number of ^{235}U peaks and the two strong uranium kα1 and kα2 x-ray peaks. The 92.365 keV and 92.790 keV ^{238}U peaks are very near the 93.456 keV Th-kα1/^{235}Upeak.

The U235 software was developed with the intent to extend the applicability of the code to very low ^{235}U concentrations and to very high ^{235}U concentration sources. Presently the code works for uranium samples that are 0.05% ^{235}U to 95% ^{235}U. Code algorithms have been found that very precisely subtract the background signal and fit the observed peak shapes. U235 is one of the codes which has been under development via a cooperative R&D agreement (CRADA) between Lawrence Livermore National Laboratory and EG&G ORTEC.

D. CZTU

A cadmium-zinc-telluride (CZT) detector is a room temperature semiconductor detector. The energy resolution of CZT is midway between that of an HPGe (high purity germanium) detector and a NaI (sodium iodide) detector.

The most serious limitation CZT detectors have is their small detector volume. This limits their ability to obtain good counting statistics in a reasonable amount of time from most uranium samples. Their peak resolution at 2.1 keV FWHM at 90 keV is adequate for a number of analytical purposes.[9-14] This resolution will always limit their accuracy to less than that of a HPGe detector with its 0.5 keV resolution at 90 keV.

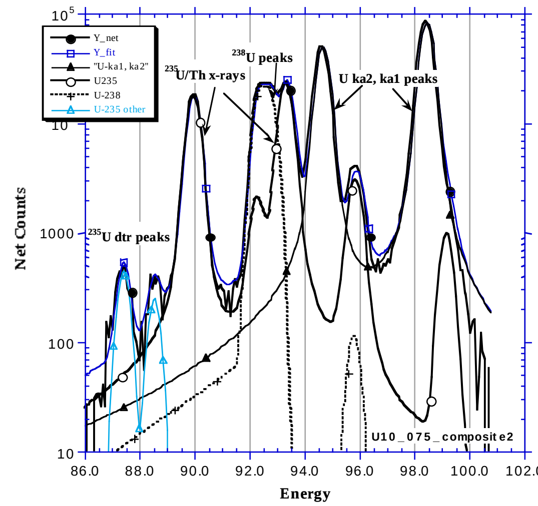

Fig. 3. The net count spectrum from 86 to 102 keV of a 10.075% ^{235}U sample with the peaks grouped into their respective components. At this ^{235}U concentration the ^{235}U and ^{238}U peaks are approximately equal. The fitting process uses both the protactinium and thorium x-rays from the ^{235}U daughters to find the best fit to the combined ^{235}U and ^{238}U spectrum.

Because of their small size, however, the CZT detectors are extremely portable.

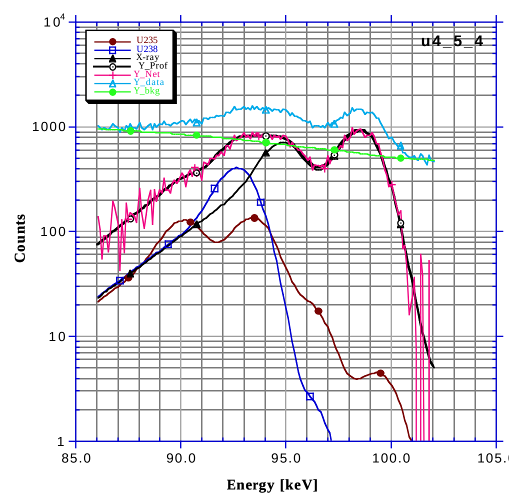

Fig. 4. The CZTU analysis for a 4.46% ^{235}U sample. The spectrum data, background, data after background subtraction, and three sources of gamma rays are shown.

As part of the MGA++ code development, CZTU analysis code was developed to provide uranium isotopic analysis for data collected with a CZT detector.[15] The CZTU code non-destructively determines the relative abundance of ^{235}U, ^{238}U and ^{234}U ratio in a uranium sample. The code uses gamma-ray spectra collected with a room temperature CZT detector. The analysis methods used are similar to those in the MGA code.

The analysis of ^{234}U depends on the 120.9 keV gamma ray. Typically, the statistics of the 120.9 keV peak are poor.

CZTU uses the 50 – 200 keV region of the gamma-ray spectrum. The code The spectrum analyzer should have a gain of approximately 105 eV/channel, with 2048 channels minimum. The 185.7 keV peak is used for self-calibration and also to confirm the presence of ^{235}U, so the gain must be set to include this peak. The sample container must be sufficiently thin to allow radiation in the 100 keV energy region to be readily observed in the spectrum. Because of the small detector size and hence the low efficiency, the collection time is typically 30 minutes. With good statistics, uranium concentrations over most of the range of interest can usually be analyzed to about ±10% accuracy.

A background determination is made within the CZTU software code. Fig. 4 shows the CZTU data and the analysis results. The top line is the original data. The flat line running through the data is the calculated fit of the background data. The dark line is the data after background subtraction.

The remaining three lines in Fig. 4 are the fits of the three main contributions to the spectral data. The method of peak fitting uses families of peaks. The four families are: ^{235}U, ^{238}U, uranium x-rays and lead x-rays. The Pb x-rays are typical contaminates that show up if any Pb collimators or shielding are near the source and detector. The uranium x-rays are produced by gamma- and alpha-produced fluorescence in the uranium source material and are always present in varying intensities. The peak shapes are described by their peak parameters, and the amplitudes are determined by branching ratios.

The two main peaks used in the83 –103 keV energy region are ^{238}U/^{234}Th 92.7 keV peak, and the ^{235}U/Th x-ray peak at 93.35 keV.

In Fig. 4, the lowest intensity line with a double peak, is the ^{235}U contribution to the gamma ray spectrum. The single peak line is the ^{238}U contribution. The largest contribution to the data (after subtraction) is from uranium x-rays. CZTU gives accurate isotopic ratios for enrichments between 0.72% and 95% ^{235}U. As can be seen in Fig. 4, very low and very high enrichments are extremely difficult to analyze because the part of the spectrum that can be clearly attributed to either ^{235}U or ^{238}U becomes very small.

E. U235HI, CZTPU and UPUHI

For situations in which the uranium is inside a thick-walled container, it is difficult or impossible to use the 100 keV energy region. We are beginning to develop a code which will analyze plutonium gamma-ray data and provide plutonium isotopic information. The U235HI code will be based on the U235 analysis code. Two sets of data are being used in the code development. The first set was provided by the International Atomic Energy Agency (IAEA) in Vienna, Austria. The second data set was collected at the Portsmouth Gaseous Diffusion Plant in Portsmouth, Ohio.

The UPUHI code will be based on the MGA and MGAHI analysis codes.

CZT detectors are limited in size, hence the energy region that will be used for the CZTPU analysis code will be limited. The usable energy range will be the same as for CZTU: 50 – 200 keV. As seen in Fig. 1, the plutonium energy spectrum in the 100 keV region is extremely complex. As in the case of CZTU, the code will rely on families of peaks.

III. DISCUSSION

We have established a strong capability for non-destructive analysis of plutonium samples with MGA. The MGA code has been expanded in to the MGA++ suite of codes. The original code, MGA, has been upgraded and licensed to EG&G ORTEC and also delivered to Euratom. The U235 code has also been licensed to EG&G ORTEC. A third code, CZTU, has been licensed to EG&G ORTEC and to Canberra.

The MGA code is currently being used at many sites in the US, in Europe and in Russia. We are currently working on a series of measurements to insure that MGA and the related codes give accurate results all possible measurement scenarios.[16]

ACKNOWLEDGEMENTS

The authors would like to thank Kenneth Raschke of Lawrence Livermore National Laboratory, and Tim Twomey, Ron Keyser and Susan Haywood of EG&G ORTEC for their contributions to the cooperative R&D for development of MGA++.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

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