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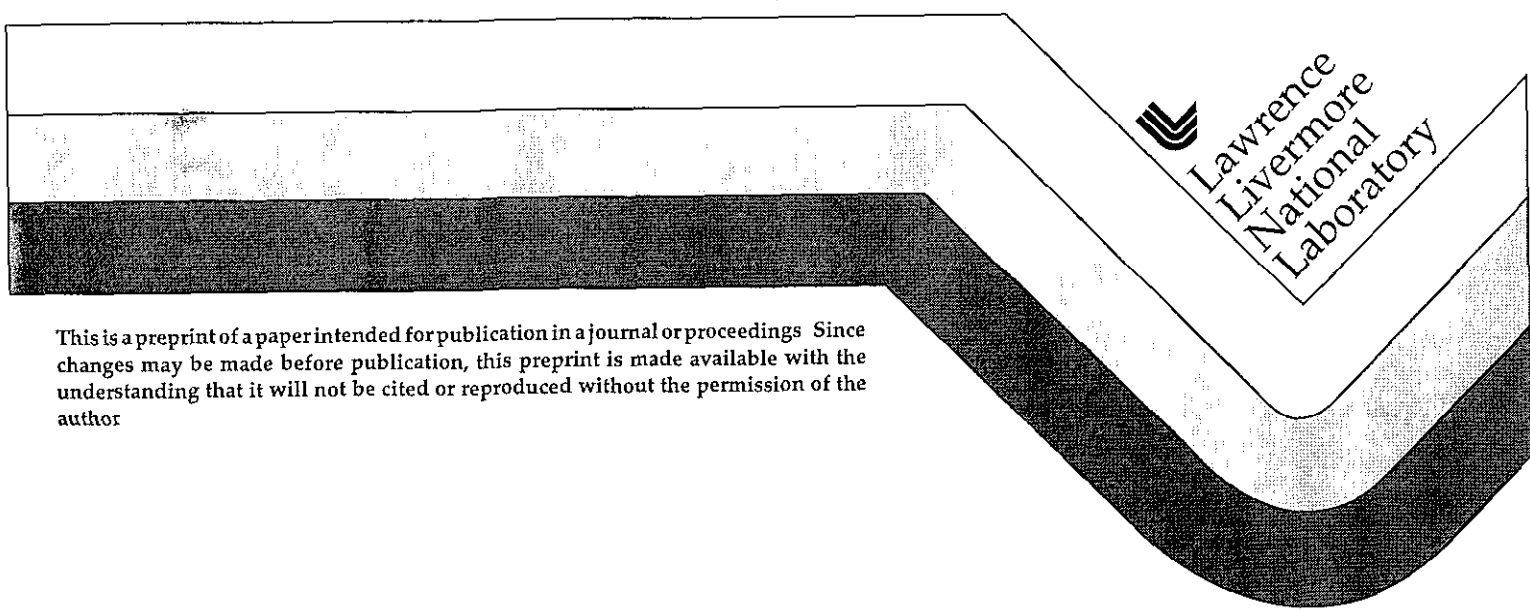

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Uranium and Plutonium Isotopic Analysis Using MGA++

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Abstract

The Lawrence Livermore National Laboratory develops sophisticated gamma-ray analysis codes for the isotopic analysis of nuclear materials based on the principles used in the original MultiGroup Analysis (MGA) code Over the years, the MGA methodology has been upgraded and expanded far beyond its original capabilities and is now comprised of a suite of codes known as MGA++ The early MGA code analyzed Pu gamma-ray data collected with high-purity germanium (HPGe) detectors to yield Pu isotopic ratios While the original MGA code relied solely on the lower-energy gamma rays (around 100 keV), 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 The MGA++ suite also includes capabilities to perform U isotopic analysis on data collected with either HPGe or cadmium-zinc-telluride (CZT) detectors These codes are commercially available and are known as U235 and CZTU, respectively A graphical user interface has also been developed for viewing the data and the fitting procedure In addition, we are developing new codes that will integrate into the MGA++ suite These will include Pu isotopic analysis capabilities for data collected with CZT detectors, U isotopic analysis with HPGe detectors which utilizes only higher energy gamma rays, and isotopic analyses on mixtures of Pu and U

Introduction

MGA++ is a suite of computer codes that analyses gamma-ray spectra and determines the isotopic abundances of actinides in a sample All data used in the MGA++ code suite is collected by non-destructive means The original MGA code was developed to determine plutonium isotopic abundances for gamma-ray data taken with germanium detectors MGA++ now includes codes designed to perform uranium isotopic analysis on data collected with germanium detectors and cadmium-zinc-telluride detectors The MGA++ code suite is fully modularized and consists of 1) an upgraded version of the original MGA code which relies on the 100 keV region, 2) MGAHI, a plutonium isotopic analysis code which uses the 200 keV - 1 MeV energy region, 3) U235, a uranium isotopic analysis code which uses gamma rays less than 300 keV, and CZTU, a uranium isotopic analysis code which uses gamma rays less than 200 keV collected with a CZT detector With the exception of CZTU, the codes analyze gamma-ray data collected with a HPGe detector All of the executable software is 32-bit and Windows 95/NT compliant

The MGA methodology, including detailed descriptions of peak shapes, efficiencies, geometry considerations, and background subtraction is described in detail in Refs 1-3 and is briefly summarized here

The number of atoms of a particular isotope is related to the observed peak intensity by the following relation

$$I_i = \lambda_i A_i B_i \Omega_i \varepsilon_i \tau_i \text{ (counts/sec)}, \qquad (1)$$

where

I_1 = measured peak intensity of isotope one,

$\lambda_1 = 0\ 6932/T_{1/2}$ = decay constant of isotope one,[4]

A_1 = number of atoms of isotope one,

B_1 = branching ratio of isotope one,

Ω_1 = fractional solid angle of the detector,

ε_1 = gamma-ray counting efficiency of isotope one, and

τ_1 = gamma-ray transmission to the detector.

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 Early work in this area[3] included development of a way to minimize the effects of these experimental problems by using the knowledge that neighboring-energy gamma rays have similar counting efficiency. That is, 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

$$A_1/A_2 = I_1\lambda_2B_2\varepsilon_2\tau_2/\ I_2\lambda_1B_1\varepsilon_1\tau_1, \qquad (2)$$

where

A_1/A_2 = isotopic ratio, and I, λ, B, ε, τ, and A are defined above

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,[5] is determined by fitting observed peak intensities to a function used to describe the relative efficiency with energy. The functions that are commonly used are adequate to determine the efficiency relationships of closely spaced peaks, *i e* those separated by less than 5 to 10 keV The intrinsic efficiency curve is used to accurately relate groups such as the 129-keV and 148-keV groups in Pu.

Analysis is greatly simplified by the following observations.

$\varepsilon_2\tau_2 / \varepsilon_1\tau_1 \approx 1$ if the two gamma rays are close to the same energy

$\Omega_1 = \Omega_2$, the fractional solid angle of detector is the same for both gamma rays

I_1 and I_2 have to be determined extremely accurately to get precise isotopic ratios

Descriptions of the codes are summarized below

MGA

MGA analyzes gamma-ray data collected with a 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 MGA also calculates the isotopic abundances of ^{241}Am, ^{235}U, and ^{238}U 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. Our current version of the MGA code has well documented and traceable changes to meet quality assurance requirements

There are about ten energy regions in a plutonium gamma-ray spectrum that can be used to calculate isotopic abundance ratios. In a few of these regions, the peaks are closely spaced, yet well resolved and isotopic abundance ratios are easily calculated. In regions where peaks are not closely spaced, a more careful delineation of a relative efficiency curve is required Finally, other regions contain several, closely spaced and overlapping peaks To properly analyze such regions, it is necessary to develop techniques for unfolding the overlapping peaks using algorithms that accurately describe the shapes of the gamma- and x-ray peaks These energy regions, ranging from 38 keV to above 1 MeV, are described extensively in Ref 3. The MGA analysis relies very heavily on the 100 keV energy region.

Although plutonium samples of less than 1 mg. can be measured, a more practical lower limit is about 100 mg. There is no upper size limit, but since the mean penetration of the gamma rays measured is very small, only the surface of a large sample is actually measured in the MGA analysis

MGAHI Development

The original MGA[3] has two data analysis modes: the one-detector mode and the two-detector mode. The one-detector mode uses a high resolution LEPS detector to collect the 100-keV gamma-ray region used to obtain the Pu isotopic information The two-detector mode uses both a LEPS and a COAX detector This COAX detector allows us to measure gamma-ray energy to 1 MeV easily With this additional information, the sample's homogeneity and isotopic content can be refined However, in the two-detector mode, the information from the LEPS detector is required, the COAX information alone is insufficient This LEPS requirement puts significant limitations when applying the original MGA code to the gamma-ray spectra of heavily shielded samples

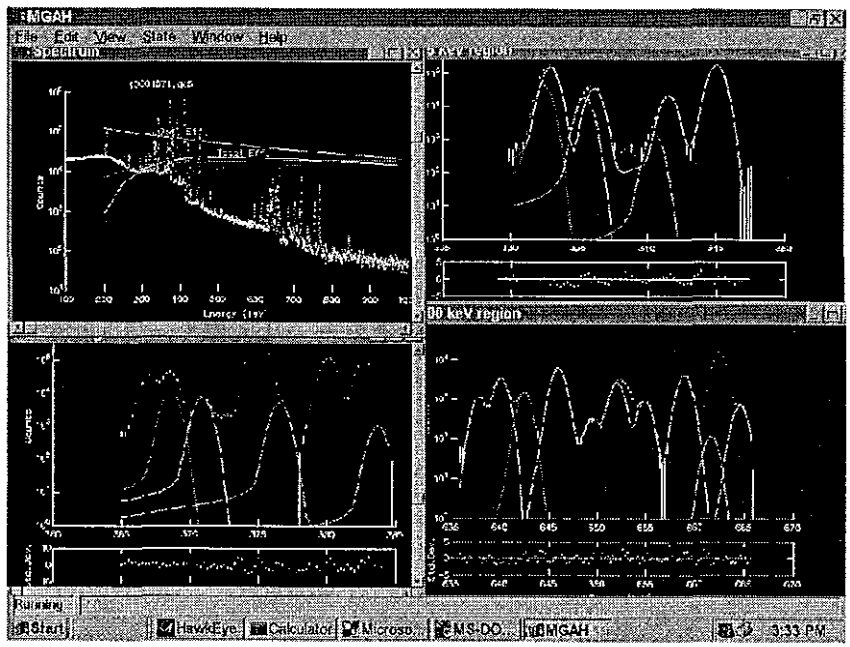
We are currently developing a spectrum de-convolution engine named MGAHI to overcome this deficiency The MGAHI uses the original MGA methodology that: 1) uses physical parameters to take into account both attenuation and emission of the gamma rays, and 2) does not require detector efficiency calibration. However, unlike the original MGA, the LEPS information is no longer required. Presently, the MGAHI requires the gamma-ray information from 200 keV to 1 MeV to determine the Pu isotopic information Once the spectrum is read into the MGAHI code, the detector efficiencies, the absorber thickness and the Pu thickness are calculated using known gamma-ray peaks from the decay of ^{239}Pu After applying the attenuation/efficiency corrections, two regions in the 300-keV and one at 600-keV were group fitted along with other discrete peaks. The final isotopic information is derived from a least-squares fit of the isotopic ratios obtained from various regions with appropriate weighting factors

Fig 1 shows a screen dump of MGAHI running in the Microsoft Windows95/NT4 0 environment A plot of the COAX spectrum that has been analyzed is shown on the top left sub-window along with the calculated total detector efficiency--the 100 keV region was completely attenuated, the first visible gamma-ray energy peak is the 203 keV line from the decay of ^{239}Pu The other sub-windows are plots of the three major group fitting regions, the residuals are also shown at the bottom of each group fitting Table 1 shows some of our preliminary results of some calibration Pu standards from the MGAHI fitting compared to their declared values

Table 1 MGAHI Pu weight % results of the two Pu (PIDIE) standards. Data were collected using a 75% COAX detector for 3 hours each Two absorbers (5mm stainless steel (ss) and 2mm Pb) were used Results from destructive analysis (DA) are also tabulated.

	^{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 +- 13%	84 91 +- 1%	14 04 +- 3%	1.01 +- 3%
PIDIE#3 DA	0.0475	84.835	14.128	0.99

Fig 1 A Screen dump of MGAHI running in the Microsoft Windows95 environment



We are also investigating the possibility of using other gamma-ray energy ranges so that the MGAHI engine can be more flexible For example, if the sample is slightly shielded the 100 - 200keV region, then perhaps the 100-keV region can also be analyzed On the other hand, if the sample is extremely heavily shielded, we may be forced to use gamma rays with energies above 300 keV (or even above 600 keV) to obtain isotopic information Of course, the narrower in the energy windows, the less information about the sample heterogeneity can be obtained

U235

A ^{235}U analysis code, U235, determines the percentage of ^{235}U, ^{238}U, and ^{234}U in a uranium sample from the analysis of the emitted gamma rays Work is now underway to improve the accuracy of the calculation, particularly at the high (>95%) and low (<0 5%)

^{235}U concentrations. A technique has been found to evaluate low ^{235}U concentrations that works well on the existing standards. Work is also proceeding on: 1) ways to better determine gamma backgrounds, 2) techniques to determine the equivalent thickness of the sample to correct for gamma attenuation, 3) evaluation of the existing data base of branching ratios of ^{235}U, ^{238}U and their daughter gamma rays to allow better results and 4) evaluation of the existing data base on the emission ratios for uranium, thorium, and protactinium x-rays [1]

^{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-ray decay, these elements decay by internal conversion, IC, and subsequent emission of daughter-product x-rays. Accurate analysis of a radioactive sample by high resolution germanium detector spectrometry requires correct information on the gamma-ray and x-ray branching ratios for the radionuclides in the sample. In addition, x-rays are also produced by gamma rays interacting (via the photoelectric effect) in the material itself—so called fluorescent x-rays. In the case of a pure uranium sample these will be uranium x-rays Internal conversion processes give rise to characteristic x-rays of the daughter product (not the parent) and are not proportional to the amount of material (the amount of thorium in a decaying sample of purified uranium is very small) in the sample Internal conversion induced x-rays are proportional to the number of decays; each decay has a fractional output of x-rays of the daughter product regardless of the parent material present in the sample This fact makes these x-rays usable for isotopic analysis if the sample has a very low concentration of daughter material (Th) To accurately use these IC x-ray peaks requires that the thorium present in very old "natural" uranium samples be removed X-rays induced by the photoelectric effect (fluorescent x-rays) have energies characteristic of the bulk material— being proportional to the mass of material present in the source. The observed x-rays, from both fluorescent and internally converted sources, must originate within a mean free path of the surface to be easily observed

Branching ratio and gamma, x-ray energy data have been published in various places[6 9] for ^{235}U and ^{238}U and some of their daughter products, details of what is used in the U235 code are found in Ref 2

There are several energy regions in the uranium gamma-ray spectra that can be used to calculate isotopic abundance ratios In this program, only gamma- and x-rays less than 300 keV are used This energy region is measured by a typical low energy Ge detector The only serious limitation this energy range imposes is the relative few ^{238}U (and ^{238}U daughters) peaks less than 300 keV Fortunately there are two relatively strong $^{238}U/^{234}Th$ lines at 92 365 and 92 790 keV and a relatively strong IC x-ray at 93 356 keV ^{235}U/Th-kα1 One of the disadvantages of using gamma rays in the 80 to 300 keV range is their limited transmission through thick material This restricts the applicability of the analysis procedures to homogenous sources or thin heterogeneous uranium sources.

CZTU

CZTU is a gamma-ray analysis program that provides isotopic analysis of ^{235}U, ^{234}U, and ^{238}U in a uranium sample from the analysis of the emitted gamma rays However, this code utilizes gamma-ray spectra measured from room temperature cadmium zinc telluride (CdZnTe or CZT) detectors

CZT detectors[10-16] have a resolution midway between that obtained with sodium iodide and germanium crystal detectors. The CZT detector's most promising feature is its ability to make measurements at room temperatures without the cryo-cooling required of HPGe detectors. The CZT detector resolution is a significant improvement over that of NaI detectors and allows much more detailed non-destructive analysis of uranium samples 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 This resolution will always limit their accuracy to less than that of a HPGe detector with its 0 5 keV resolution at 90 keV.

CZTU uses the 50 to 200 keV region of a gamma-ray spectrum of a uranium sample to calculate the ^{235}U isotopic enrichment The code utilizes the ^{235}U peak at 185 715 keV for self-calibration and confirmation purposes, hence the gain should always be set to at least include this peak The sample container must be sufficiently thin to allow radiation in the 100 keV energy region to be readily detectable

As with the other MGA++ codes, the method of peak fitting utilizes families of peaks whose shapes are described by their peak parameters and whose amplitude is determined (after corrections) by their branching ratio The five groups of sources are: ^{235}U, ^{238}U, U x-rays, Th x-rays and Pb x-rays These groups can be further divided into four families ^{235}U + Th x-rays + Pa x-rays + daughters, ^{238}U + Pa x-rays + daughters, U fluorescent x-rays and Pb fluorescent x-rays.

The Pb x-rays are typical gamma-ray "contaminates" that show up if any Pb collimators or shielding are near the source and detector. These gamma rays are produced by gamma-ray induced fluorescence in the lead These peaks complicate the analysis and should be minimized whenever possible.

The U x-rays are produced by gamma and alpha produced fluorescence in the uranium source material and are always present in varying intensities X-rays are also produced by internal conversion processes that are characteristic of the daughter, not of the parent These x-rays are proportional to the number of decays and have known branching ratios

The results indicate that most CZT spectra can be analyzed for ^{235}U concentration to about ±10% average accuracy for samples with reasonable statistics - greater than $2x10^6$ total counts in the spectra This uncertainty increases at both low (<1%) and high (>50%) ^{235}U concentrations ^{234}U concentrations, because of the poor statistics in the 120 9 keV peak, can only be determined, for some samples, to about ±50% accuracy On occasion, a single analysis gives much larger than ±10% uncertainty These fluctuations are connected with statistics in determining the background correctly and improve with better counting statistics

Further code development is needed to develop more stable algorithms for background, peak determinations and efficiency/transmission corrections

Graphical User Interface

We have developed a prototype graphical user interface (GUI) for the CZTU code This user interface is easily extensible to MGA, MGAHI and U235 However, unless there is strong customer interest in doing this, we will probably defer further GUI deployments to commercial licensees Canberra provides a GUI in its U/Pu software EG&G Ortec provides MGAView, U235View and CZTView GUI software for the analysis codes

We have also developed a graphics server, SpecView, for viewing spectral data and graphical representations of region fit results from any of our codes SpecView is a highly flexible client-server architecture which allows a number of executing analysis programs to draw an unlimited number of "standard views" of the data simultaneously This has proven particularly useful in that it allows code developers to trace the progress of a complex fitting process visually The interface to this server is independent of the computer language used in the analysis product as well as the number and type of networked clients requesting the drawing service The program itself is designed from the ground up as an object oriented C++ application fully utilizing Microsoft's latest operating system technology

The commercial versions of the analysis codes from EG&G Ortec will be supplied with graphics capabilities

Conclusions

The MGA, U235 are fully developed and have been licensed to EG&G Ortec CZTU is also fully developed and has been licensed to EG&G Ortec and to Canberra The MGAHI code is in the alpha-test phase of development We continue to push the frontier of gamma-ray spectroscopy of SNM, and we are developing new codes which we will integrate into the MGA++ suite Further work will include development of a code that performs plutonium isotopic analysis for gamma-ray data collected with CZT detector Our uranium isotopic analysis capabilities will be extended to include a code which will utilize gamma rays in the range of 200 keV to 1 MeV and will be used to analyze uranium samples that are enclosed in thick walled containers

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