

Advanced Gamma-Ray Isotopic-Ratio Actinide Analysis from Ge Detector Spectra

- A Suite of Three Advanced Analysis programs:
 - MGA analyzes Pu with a single planar, or one planar and one coaxial Ge detector
 - MGAHI analyzes Pu with a single coaxial Ge detector
 - U235 analyzes U only with a planar Ge detector
- The latest LLNL algorithms for actinide analysis
- No calibration standards necessary to correct for matrix or container effects
- Rapid operation with display of spectra, results, and peak-fit residuals
- Easily modified parameter sets for optimum results
- Flexible reporting: Instant results and archive copy to Access® database
- Integrated data collection and analysis
- Easy-to-use Windows® Graphical User Interface
- ORTEC CONNECTIONS-32 compliant
- Operates with all ORTEC and many non-ORTEC MCAs
- Developer's toolkit options aid custom system development

MGA++ is a suite of three software programs (MGA, U235 and MGAHI) for analysis of Actinide spectra acquired by germanium detectors. MGA++ is the result of years of continuing development at Lawrence Livermore National Laboratory.¹

The original MGA code was developed to determine plutonium isotopic abundances for gamma-ray data taken with germanium detectors. MGA-B32 consists of 1) an upgraded version of the original MGA code, which relies on the 100-keV region; 2) U235, a uranium isotopic analysis code that uses gamma rays less than 300 keV; and 3) MGAHI, a plutonium isotopic analysis code that uses the 200 keV–1 MeV energy region. The codes analyze gamma-ray data collected with a HPGe detector. All of the executable software is 32-bit Windows compliant.

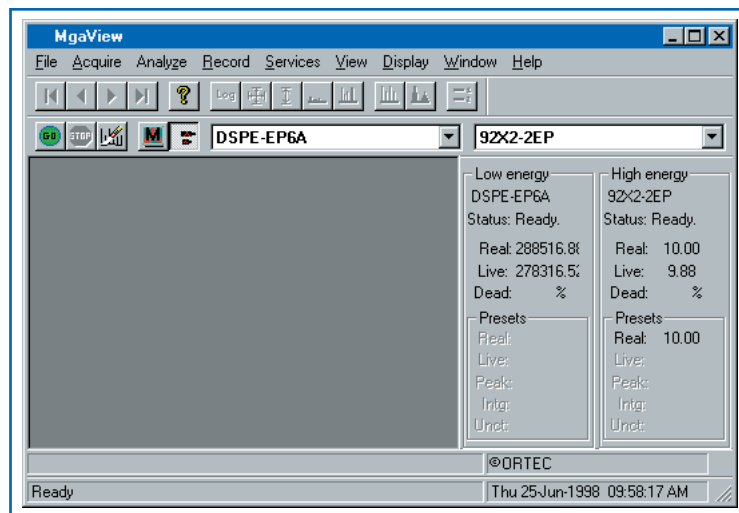
The programs, requiring no special calibration sources or calculations, use only information obtained from the sample spectra to determine the isotopic ratios.

A full member of the CONNECTIONS-32 software family, MGA++ can display the data being acquired, then rapidly analyze and report results — all from a single, easy-to-use program.

User Interface

For each operational mode, separate “viewer” programs (MGAView, MGAHIView and U235View) and analysis modules (MGA.EXE, MGAHI.EXE and U235.EXE) ensure the integrity of the analytical methods. The Viewer program provides the user interface and the hardware control function. In MGA++ it is possible to suppress all spectral display.

MGAView, MGAHIView and U235View present the same friendly user interface. The MGA mode operator interface is shown in Fig. 1. The current status of the detector is shown on the right. A special count-rate meter mode shows the instantaneous count rate of a selected region. The spectrum may be viewed during acquisition.



The Start/Save/Report feature (Fig. 2) provides one-button collection and analysis — via keyboard or mouse — easing the task for operators in protective suits! When additional counts are deemed necessary, “Restart/Save/Report” continues the current count.

Reanalysis of spectra from disk or in the MCB is easy — just click and select (Fig. 3). All the spectra collected by the MGA++ programs are stored in the ORTEC standard spectrum (SPC) format, which can be read by many programs. The file includes analysis parameters and hardware description records — everything needed to verify the results.

Spectra stored on disk may be analyzed by MGA++ from a wide variety of input file formats in addition to the primary ORTEC SPC file format (Table 1).

The analysis results can be displayed, with zoom function capabilities (Fig. 4), for visual confirmation of the analysis.

Peak fit results may be scrutinized along with fit residuals for visual assurance of analysis quality. To aid comprehension, individual peaks on the display can be turned on or off (Fig. 5).

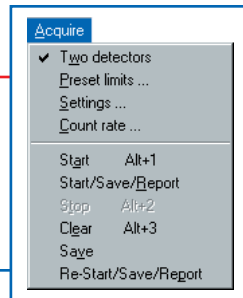


Figure 2.

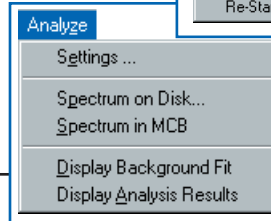


Figure 3.

File Formats Read

LLNL/ACCUDUMP (NCD BINARY)
 ASCII (sequential integer) no header
 Nuclear DATA μ MCA (ACCUSPEC) (.CNF)
 Canberra S100
 ORTEC ADCAM (.CHN)
 LLNL ASCII
 SPE ASCII
 “Euro” ASCII
 ORTEC MGA (Integer .SPC)
 ORTEC (Real .SPC)

Table 1.

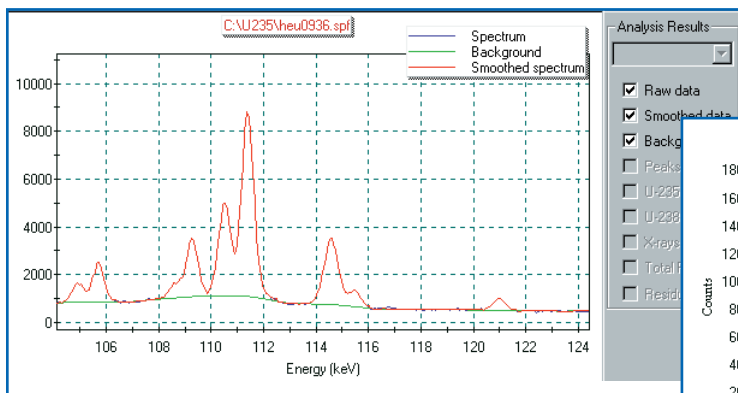


Figure 4.

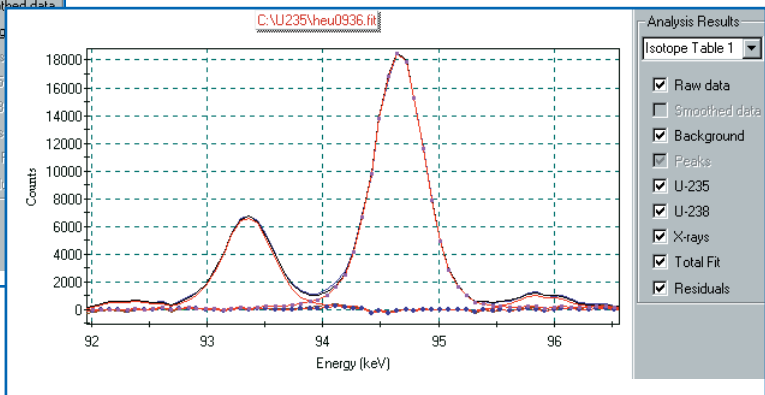


Figure 5.

Flexible Analysis

Analysis parameters are specified in clear, easy-to-understand dialog boxes. Analysis parameters may be saved to disk for use on similar samples and recalled as required.

The flexibility of the analysis settings options for U235View are shown in Figs. 6–9, for sample type, peak shape parameters, absorption, and source/detector absorption. For MGAView (Figs. 10–13), they are sample type, source geometry, ^{242}Pu calculation, and low-energy detector. Figures 14–16 show sample type, geometry, and Pu-242 dialogs for MGAHView.

At the end of each analysis the results are automatically stored in the Access format database and are also printed or displayed for the operator. The three database tables (Analysis, Acquisition, and Isotope; see, for example, Fig. 17) can be viewed by MGAView (using the record function) or by Access. This powerful data storage method makes summary reports, exception reports, and other useful outputs which are easy to create. The standard reporting options are shown in Figs. 18, 19 and 20.

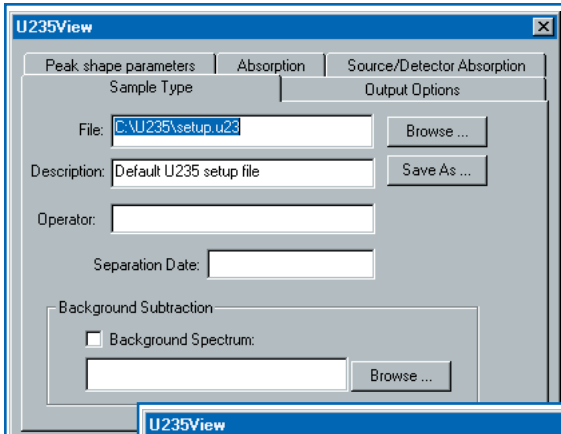


Figure 6.

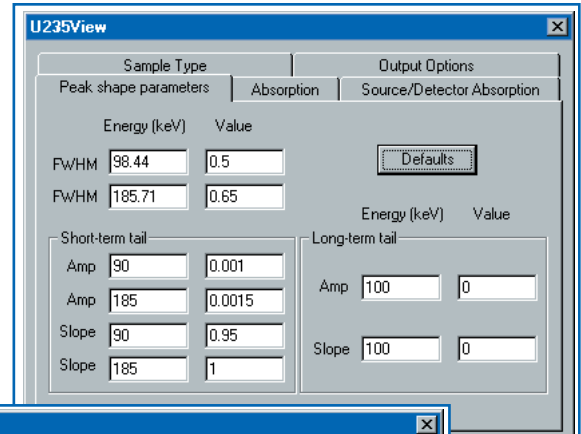


Figure 7.

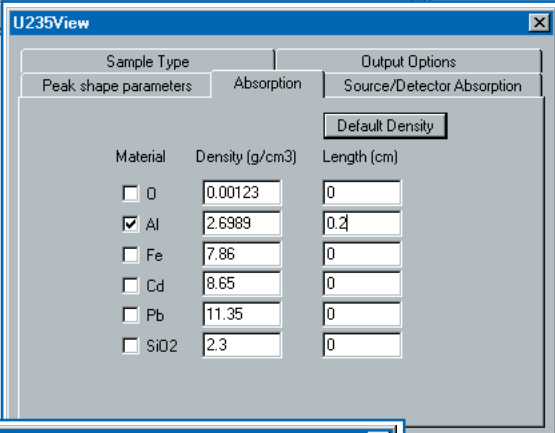


Figure 8.

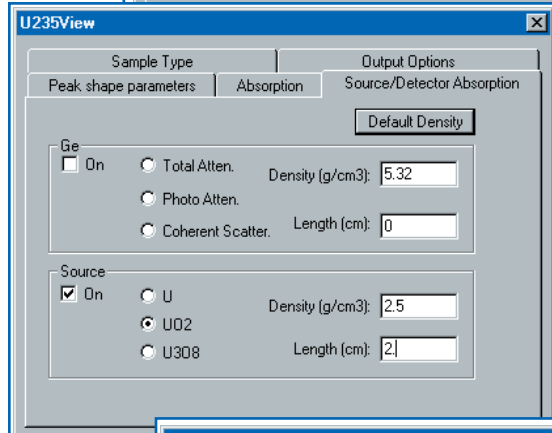


Figure 9.

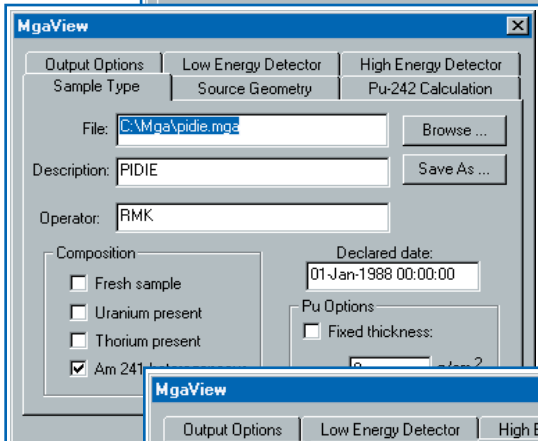


Figure 10.

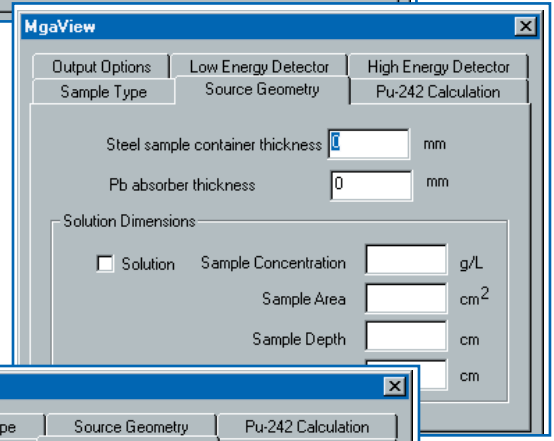


Figure 11.

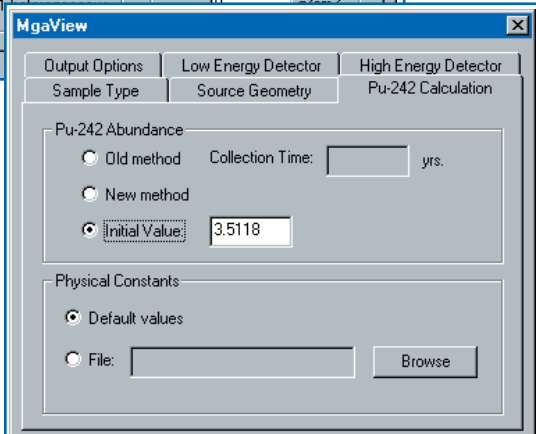


Figure 12.

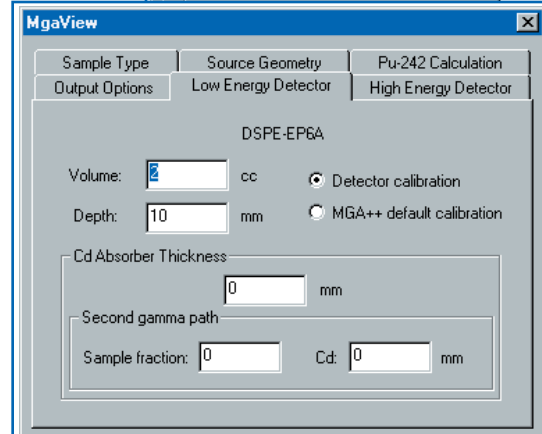


Figure 13.

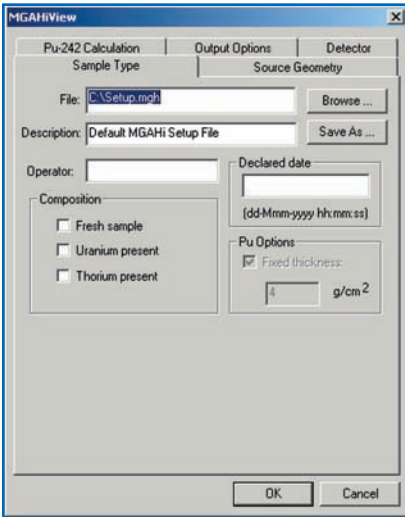


Figure 14.

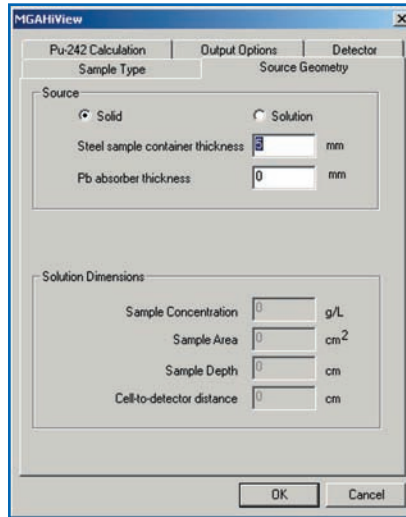


Figure 15.

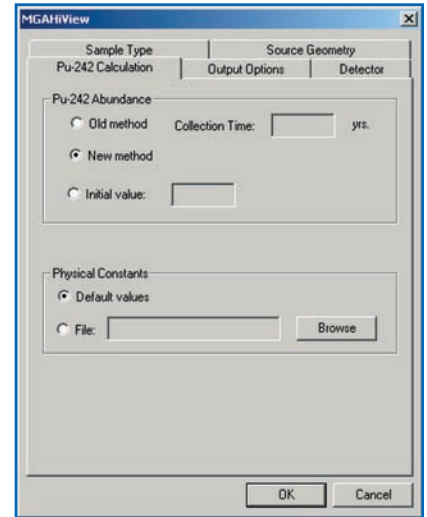


Figure 16.

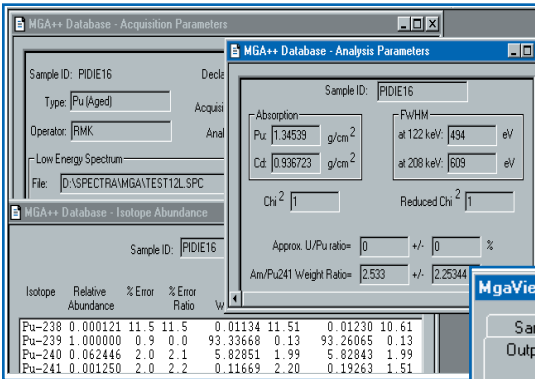


Figure 17.

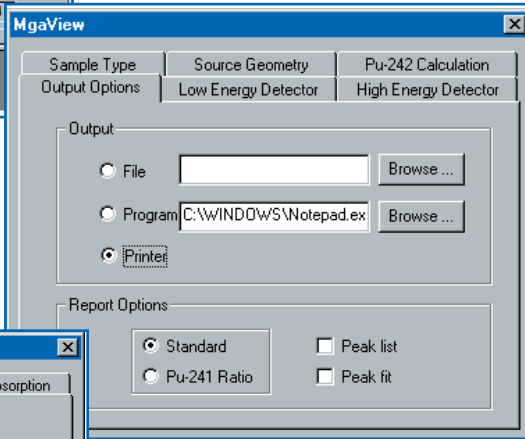


Figure 18.

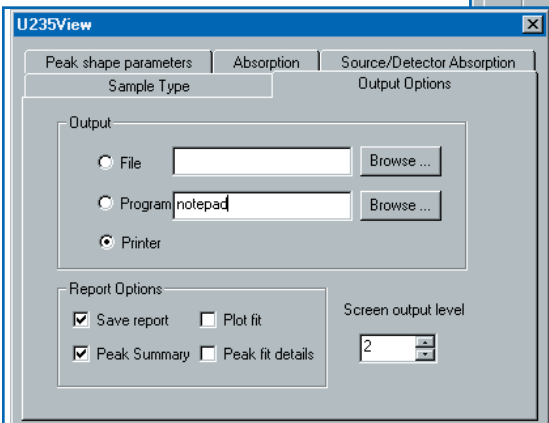


Figure 19.

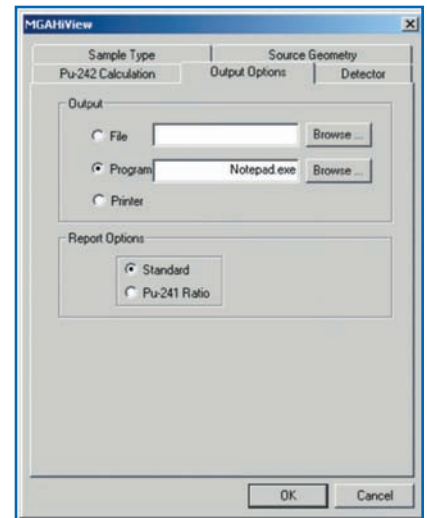


Figure 20.

CONNECTIONS Integration

The MGA++ programs, operating under Windows 2000/XP, are completely integrated into the ORTEC CONNECTIONS-32 environment. All ORTEC multichannel buffer hardware, including the DigiDART, DSPEC, DSPEC Plus and DSPEC jr MCAs, are supported — whether networked or standalone.

The CONNECTIONS structure, common to all 32-bit ORTEC application programs, facilitates development of custom applications, e.g., automated measuring systems. (Developer's toolkits are available from ORTEC to aid this process.)

Common benefits of all CONNECTIONS 32-bit products are multitasking, multi-threading support for concurrent analysis processes under Windows 2000/XP, simultaneous support of both locally connected and remotely connected MCAs over Ethernet links, and detector locking by security password.

Prerequisites

MGA++ will operate correctly on any system supporting ORTEC multichannel buffer hardware under Windows 2000/XP. While MGA++ can control compatible MCA hardware directly, MAESTRO-32 is a prerequisite for instrument setup.

MGA Analysis Mode

- Produces weight percent results for $^{238,239,240,241}\text{Pu}$, $^{241,243}\text{Am}$, $^{237,239}\text{Np}$, and $^{235,238}\text{U}$
- Determines ^{242}Pu
- Operates with a single-planar HPGe detector for 0–300 keV or with a planar and a coaxial HPGe detector for 0–1000 keV
- No calibration standards needed to correct for matrix or container effects
- Automatic energy and peak shape recalibration

MGA mode has two data analysis configurations: one-detector mode and two-detector mode. In one-detector mode, MGA is designed to operate with a planar HPGe detector for the energy range 0–300 keV to obtain the Pu isotopic information. In two-detector mode, intended for highly-attenuated samples, the measurement may be aided by higher energy data up to ~1000 keV obtained from a coaxial HPGe detector. This coaxial detector allows measurement of gamma-ray energies to ~1 MeV. With this additional information, sample homogeneity and isotopic content can be refined. However, in the two-detector mode, the information from the planar detector is required, the coaxial information alone is insufficient. This requirement adds significant limitations when applying the original MGA code to the gamma-ray spectra of heavily shielded samples. This has to some extent been mitigated by the introduction of the ORTEC SGD GEM detectors, which in some cases can be used to perform the function of the planar and coaxial detector in one, by the use of two separate spectra, low and high energy.

There are about ten energy regions in a plutonium gamma-ray spectrum which may be used to calculate isotopic abundances. Those in the 94–104 keV region and the peaks at 129 and 148 keV are the most intense. Although those regions are difficult to analyze, yield precision of 1% or better can be achieved.

Typical MGA Mode Performance



| Comparison of MGA Analysis Results, Measured at LLNL with PIDIE Standards–10 Minute Count | | | |
|---|-----------------------|----------------------|----------------------------|
| Standard | Std Std Wt % Declared | Std-Calc MGA % Error | Reported MGA % Uncertainty |
| ^{238}Pu | | | |
| 1 | 0.0101 | 0.7 | 3.87 |
| 3 | 0.0437 | 3 | 1.74 |
| 5 | 0.1221 | 3 | 1.39 |
| ^{239}Pu | | | |
| 1 | 93.848 | 0.064 | 0.07 |
| 3 | 84.927 | 0.024 | 0.08 |
| 5 | 76.534 | 0.24 | 0.18 |
| ^{240}Pu | | | |
| 1 | 5.987 | 1.02 | 0.6 |
| 3 | 14.191 | 0.056 | 0.49 |
| 5 | 21.381 | 0.9 | 0.61 |
| ^{241}Pu | | | |
| 1 | 0.1206 | 0.08 | 0.65 |
| 3 | 0.6049 | 1.6 | 0.42 |
| 5 | 1.26 | 0.8 | 0.49 |
| ^{241}Am | | | |
| 1 | 0.3048 | 0.9 | 0.39 |
| 3 | 1.012 | 0.2 | 0.38 |
| 5 | 2.55 | 0.27 | 0.49 |
| Total Specific Power | | | |
| Standard | Declared | Std-Calc MGA % Error | |
| 1 | 2.644 | 0.22 | |
| 3 | 4.068 | 0.12 | |
| 5 | 6.639 | 0.09 | |

Sample Age Since Processing

MGA handles both freshly-separated and aged samples — important because recently-processed plutonium samples are usually very low in ^{237}U and ^{241}Am . The ^{237}U increases in activity for about 2 months after processing, at which time it reaches equilibrium with the alpha-decay rate of its ^{241}Pu parent. Any analysis method that always assumes an established decay equilibrium between these two isotopes cannot be used on freshly-processed samples.

Presence of Other Radioactive Materials

Other radioactive materials may be present in plutonium samples as decay products, as contaminants from previous processes, or as a result of blending. Uranium is one common radioactive material which is blended with plutonium to form mixed-oxide (MOX) fuels. MGA can accurately determine the relative abundance of ^{235}U in a sample. Other radioactive materials sometimes encountered are ^{237}Np - ^{233}Pa , ^{243}Am - ^{239}Np , and low levels of some fission products such as ^{95}Zr - ^{95}Nb and ^{137}Cs . If a two-detector system is employed, ^{237}Np can be detected down to about 50 ppm by analysis of the 312 keV peak of ^{233}Pa , the daughter of ^{237}Np . MGA automatically recognizes the presence of interference from ^{243}Am and ^{239}Np isotopes, the fluorescence of x-rays of thorium in the sample, and the presence of ^{137}Cs (if the two-detector system is used).

MGAHI Mode

MGAHI uses physical parameters to take into account both attenuation and emission of 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 high background, space-limited environment. With too much shielding, the 100-keV energy region could be completely attenuated. In a high-background environment, the detector could see gamma rays from other nearby sources. Also, the spectrometry system could have a very high dead time. Lead shielding can be used in this situation to reduce the dead time, but this could also cut out the low-energy gamma rays. MGAHI employs the original MGA methodology in that it: 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 planar information is no longer required. The MGA analysis relies very heavily on the 100 keV energy region detector efficiencies, absorber thickness and Pu thickness are calculated from the spectral data using known gamma-ray peaks from the decay of ^{239}Pu .

MGAHI Mode Performance

MGAHI Pu weight % results of the two Pu (PIDIE) standards. Data was collected using a 75% coaxial detector for 3 hours. Two absorbers (5 mm stainless steel (ss) and 2 mm 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 |

U235 Analysis Mode

- Relative ratios for ^{235}U , ^{234}U , and ^{238}U
- Automatically checks for the presence of Pu using the 129 keV peak
- Operates with a single-planar HPGe detector from 0–300 keV
- No calibration standards necessary to correct for matrix or container effects
- Corrects for internal sample absorption and absorbers placed between sample and detector
- Peak shape calibration determined from user-defined spectral peaks, reloadable from file, or from default values

U235 uses a single planar HPGe detector and operates in the energy range 0–300 keV. It determines the relative isotopic ratios for Uranium. In a similar fashion to MGA and MGAHI, no primary calibration is required for efficiency or for absorbers in the matrix or in the sample container.

Typical U235 Mode Performance

U235 gives weight percent results for ^{234}U , ^{235}U , ^{238}U .

| Isotope | Range (wt. %) | Absolute Accuracy (%) |
|------------------|---------------|-----------------------|
| ^{234}U | 0.02–2 | 5.0 |
| ^{235}U | 0.02–0.5 | 5.0 |
| | 0.5–70 | 0.5 |
| | 70–93 | 1.5 |
| | 93–99 | 5.0 |
| ^{238}U | 99–99.5 | 5.0 |

The U235 analysis mode gives relative ratios for ^{235}U , ^{234}U , and ^{238}U ; and when operating in this mode, it can warn the user of the possible presence of Pu.

Absolute accuracy depends on the statistics of the sample.

General Considerations

Sample Size and Matrix

In practical terms, the lower limit on sample size is about 100 mg — determined by count time and required statistical accuracy. While in principle there is no upper limit to the size, only the surface of a large sample is actually measured - because plutonium and uranium have short mean-free-paths for low-energy photons. Increasing the sample thickness beyond the “saturation thickness” will not increase the count rate because photons from the back of the sample are fully absorbed.

The extent of fluorescence from uranium or plutonium by their own alpha or gamma emission is much lower than from solid materials. Entering details about the solution will assist the analysis in dealing with the x-rays.

Shielding

A shield or collimator should be used to reduce or eliminate the counts in the spectrum from nearby materials. Cadmium, copper, lead, or tantalum foils are used to reduce the presence of the low-energy gamma rays, specifically those at 59 keV. This reduces the dead time from unusable counts for plutonium samples. These low-energy gamma rays are hardly ever present in uranium samples. Unless they are present, preferential filtering of low-energy photons is almost never required and is of no value.

Germanium Detector

For measurements up to 300 keV with either MGA or U235, a planar HPGe detector with a nominal resolution of 550 eV at 122 keV is required. While this requirement may in some cases be relaxed, the 122-keV resolution should not exceed 700 eV.

A thicker planar detector will have a higher peak-to-Compton (p/C) ratio, so that, assuming that it has an area and 122-keV resolution consistent with the other requirements specific to the measurement, it may deliver superior performance at low energies.

To use the higher energy regions in the analysis of samples containing only plutonium, in two detector MGA mode or in MGAHI mode, a coaxial detector with a resolution of less than 2.0 keV at 1332 keV is adequate. For MGAHI, a detector resolution of 1.1 keV or better at 208 keV is required. The 203-keV gamma ray from ²³⁹Pu must be visible. Up to 5 mm of Pb absorber can be placed in front of the detector.

ORTEC's Safeguard (SGD) Series of coaxial and planar detectors, specifically optimized for the purpose, are highly recommended for use with MGA++.

Ordering Information

To order, specify:

| Model | Description |
|---------|---------------------------|
| MGA-B32 | MGA++ Safeguards Software |
| MGA-U32 | Update for MGA-B32 |
| MGA-G32 | Documentation for MGA-B32 |

¹It is the subject of a cooperative research and development agreement (CRADA TSV-1368-96) and license (License Number TL-1375-96) between ORTEC and the University of California under which ORTEC is integrating those programs into the ORTEC software environment to enhance usability.

²K. Debertin and R.G. Helmer, "Gamma- and X-Ray Spectrometry with Semiconductor Detectors," p. 185, Elsevier Science Publishers B.V., The Netherlands, 1988.

ORTEC Signs MGA++ Licensing and Development Agreement with LLNL

ORTEC and Lawrence Livermore National Laboratory have entered into licensing and development agreements in the area of specialized software for nuclear safeguards applications. The license covers the existing versions of the MGA ++ suite of codes which analyze HPGe detector gamma-ray spectra from a wide variety of nuclear safeguards samples in order to determine relative plutonium isotopic abundances and/or uranium enrichment. The development agreement (CRADA) relates to further improvements to the code and an improved "seamless" integration within the ORTEC CONNECTIONS Windows 2000/XP spectroscopy environment for maximum ease-of-use and reliability.

Specifications subject to change
030609

ORTEC[®]

www.ortec-online.com

Tel. (865) 482-4411 • Fax (865) 483-0396 • ortec.info@ametek.com
801 South Illinois Ave., Oak Ridge, TN 37831-0895 U.S.A.
For International Office Locations, Visit Our Website

AMETEK[®]
ADVANCED MEASUREMENT
TECHNOLOGY