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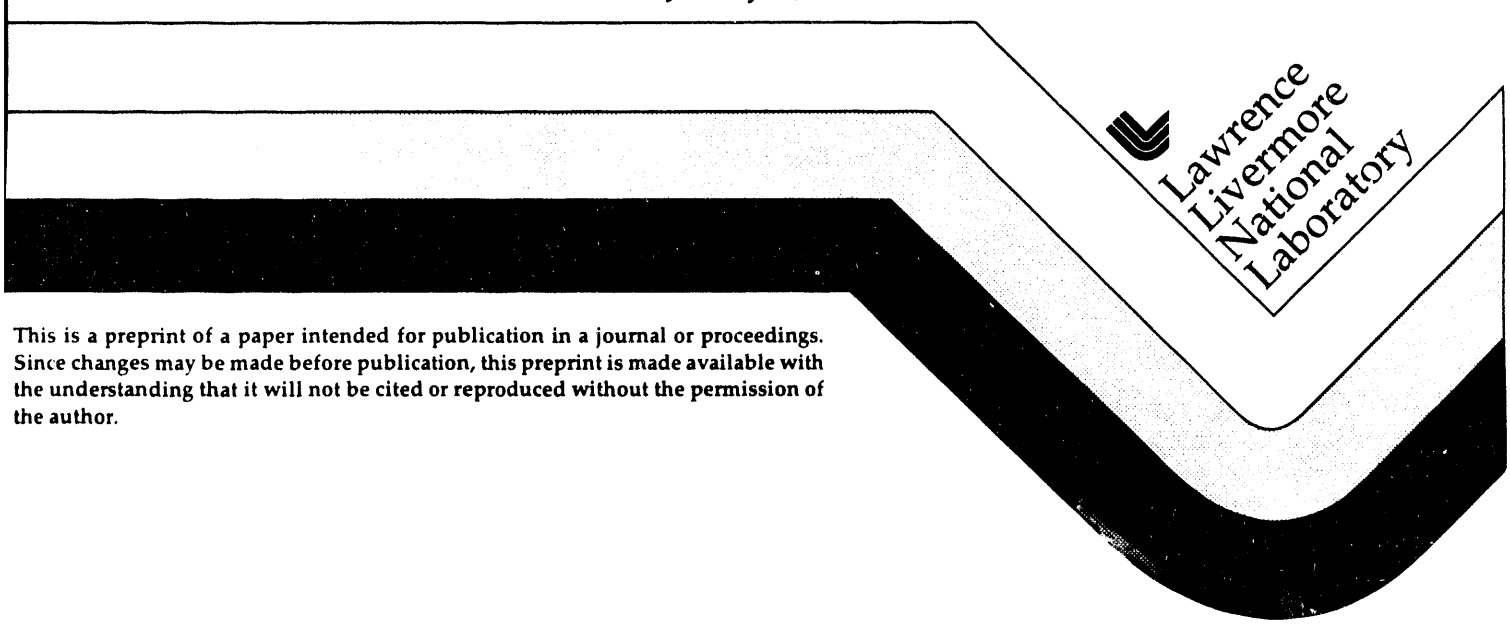

MGAU: A NEW ANALYSIS CODE FOR MEASURING U-235 ENRICHMENTS IN ARBITRARY SAMPLES[1]

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Abstract

We have developed a computerized analysis method for determining ^{235}U enrichment in uranium items where no suitable reference standards exist or where nonreproducible conditions make calibration impossible. The method requires no calibrations and is capable of accuracies of 1–2% in only a few minutes.

1. Introduction

The traditional method used for ^{235}U enrichment measurements, developed more than 20 years ago [1], is based on the measurement of the 186-keV peak of ^{235}U. Further developments of this method have been described (e.g., Refs [2 and 3]). Although simple NaI spectrometer systems can be used to make this measurement, it suffers in accuracy, requires a calibration for each new sample container, and can be used only for samples with thicknesses of several mean free paths at 186 keV. These limitations therefore tend to reduce safeguards measurements simply to consistency checks.

Although the 186-keV peak of ^{235}U is the most intense peak from the decay of this isotope, other radiations are associated with the decay of this isotope or one of its daughter products [4,5]. Hagenauer [5] describes three energy regions in Ge-detector spectra that are available for determining ^{235}U enrichments: the 53- to 63-keV, 90- to 94-, and 186- to 1001-keV regions. Although the 186- and 1001-keV peaks are easy to measure, it is difficult, because of the large difference in their energies, to determine the relative efficiencies with which they are detected. The low-energy gamma rays in the 53- to 63-keV region, which are severely attenuated by even modest amounts of attenuating materials, also are usually not useful. For these reasons, we use the radiations in the second region, which we have expanded to include both gamma and x rays from 84 to 130 keV.

An advantage in analyzing the second energy region is that the ^{235}U and ^{238}U radiations are very close in energy and therefore are detected with comparable efficiencies. Until now, the complex structure of this close energy spacing also has been the greatest obstacle to accurate analyses. However, from our experience in developing the MGA code [6] for plutonium isotopic analysis, we can unfold complex, overlapping peak groupings with relative ease.

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2. Summary of Method

The usual method of determining relative isotopic abundances from gamma-ray spectra containing peaks from two or more isotopes is to measure and interpret peak intensities of neighboring gamma-ray peaks associated with the different isotopes. The small differences in the detection efficiencies of the measured peaks usually can be determined from other information in the spectrum. Relative isotopic abundances then can be calculated by using these relative efficiency factors with known gamma- and x-ray branching intensities and half-lives for the isotopes.

This method becomes slightly more complicated when analyzing peaks that severely overlap each other. Then, the peak intensities must be determined by using peak-fitting techniques. Sometimes, the spacing of peaks is so close that ordinary fitting techniques will not work. (That is, techniques that allow the peak positions and amplitudes to be independently free in the fitting process will not converge properly.) To overcome this difficulty, we developed a "response function" method that allows many peak parameters to be fixed relative to each other [6,7] thus greatly reducing the degrees of freedom in the fitting process. In the limiting case, the resulting equations can even be made linear, thereby avoiding the iterative methods usually used. When these same techniques are applied to the peaks in the 89- to 100-keV region of a uranium spectrum, analyses can be made quickly and accurately. Note that (1) the gamma and x rays present in this region require different algorithms to describe their peak shapes, and (2) many of the peaks are due to daughter products of ^{235}U and ^{238}U, rather than directly to these two isotopes. Finally, the 94- and 98-keV peaks in this region are due primarily to self-induced fluorescence of the uranium sample.

The 89- and 93-keV x-ray peaks from the decay of ^{235}U and the 92.367- and 92.792-keV gamma-ray doublet from ^{238}U-^{234}Th are very close in energy (see Fig. 1). Therefore, this region is ideally suited for determining the relative abundances of ^{235}U and ^{238}U. Although the peaks are close, one must nonetheless determine the relative efficiencies with which the various peaks in this region are detected. This can be done easily by comparing the measured intensities of the K_α and K_β x-ray peaks of uranium, which are invariably present due to self-induced fluorescence of the sample. The 98- and 186-keV peaks are used to determine the approximate energy-scaling and peak-shape characteristics for a given spectrum.

The presence of ^{234}U also usually can be detected by the presence of its gamma-ray peak at 121 keV.

3. Results

The methods summarized here have been incorporated into a computerized analysis program called MGAU. First, a small setup file is required to describe a few system and measurement conditions (e.g., approximate gain, zero offset, and spectrum format). Thereafter, only the spectrum file name is needed to perform an analysis. To evaluate the applicability and accuracy of the new method implemented in MGAU, we acquired spectra of various standards at several different facilities. Table I contains results obtained for measurements made on 45 different samples of known ^{235}U enrichment.

The measurements were taken under various conditions. Some detectors had very good resolution, whereas others had a FWHM resolution in excess of 700 eV at 122 keV. The samples differed in size and chemical composition and came in various types of containers. Measurement times, counting rates, and shielding arrangements also varied. Perhaps the largest variable was the ^{235}U isotopic abundance, ranging from 0.017% to 97%.

The following conclusions can be drawn from these measurements:

- ^{235}U enrichment measurements can be made with accuracies of a few percent for enrichments ranging from depleted to 93% enrichment.
- When the ^{235}U abundance is about 10%, the ^{235}U and ^{238}U peaks are about equal in intensity. We may reasonably conclude, therefore, that the most accurate measurements will be

made near this abundance. We can also infer that measurement precision becomes worse near the extremes of the enrichment scale (i.e., depleted vs 93%).

- The accuracy of the analysis results appear to be nearly independent of all system, measurement, and source variables. However, the method has some limitations.

4. Limitations

Because the 92.367- and 92.792-keV gamma-ray doublet measured by MGAU results from the decay of ^{234}Th, the first limitation is that ^{234}Th activity in a sample must be in equilibrium with the activity of its ^{238}U parent. Because ^{234}Th has a half-life of 24.1 days, this equilibrium is not achieved until about 5 months after its separation from uranium.

A second limitation applies to measuring cylinders containing UF_6. The problem is that uranium and/or its thorium daughter products can form deposits on the walls of the container [8]. If this surface deposit is not carefully removed between fillings, enrichment measurements of a cylinder of UF_6 may be compromised. In one MGAU measurement that yielded obviously low analysis results, a spectrum of an "empty" cylinder displayed the 92-keV doublet peak for ^{234}Th but no peaks for ^{235}U or its daughter products. We concluded that thorium daughter products were retained from the previous filling of the cylinder. The ^{231}Th daughter of ^{235}U decayed quickly (with a half-life of 25 h), leaving only the longer-lived ^{234}Th.

The thickness of the sample container also can limit the accuracy of an analysis. The 16-mm wall of a UF_6 cylinder attenuates the 90- to 100-keV radiations by about a factor of 250. The precision of the results obviously are reduced when the signals are so highly attenuated.

5. Conclusions

Our MGAU code, which runs on a personal computer, can measure ^{235}U enrichments that are accurate to 1 or 2% by analyzing the 89- to 98-keV region of a uranium gamma-ray spectrum. Applications are limited only by the constraints listed herein and by the number of counts in a spectrum required to achieve the desired precision.

6. References

[1] REILLY, T.D., WALTON, R.B., PARKER, J.L., A-1 Progress Report, report LA-4605-MS, p.19, Los Alamos National Laboratory, Los Alamos, NM (1970).

[2] AUGUSTON, R.H., REILLY, T.D., Fundamentals of Passive Nondestructive Assay of Fissionable Material, report LA-5651-M, Los Alamos National Laboratory, Los Alamos, NM (1974).

[3] MATUSSEK, P., Accuate Determination of the ^{235}U Isotope Abundance by Gamma Spectrometry, A User's Manual for the Certified Reference Material, EC-NRM-171/NBS-SRM-969, report KfK 3752, Kernforschungszentrum Karlsruhe, Karlsruhe, Federal Republic of Germany (1985)

[4] DRAGNEV, T.N., DAMJANOV, B.P., Methods for Precise Absolute Gamma-Spectrometic Measurements of Uranium and Plutonium Isotopic Ratios, report IAEA-SM-231/130, p. 739-752 (Proc. Int. IAEA Symp., 1978), IEAE, Vienna, Austria (1978)

[5] HAGENAUER, R., Nondestructive Uranium Enrichment Determination in Process Holdup Deposits, Nucl. Mater. Manage. **XX** (Proceedings Issue), 74–78, 1991.

[6] GUNNINK, R., MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, report UCRL-LR-103220, Lawrence Livermore National Laboratory, Livermore, CA (1990).

[7] GUNNINK, R., RUHTER, W.D., NIDAY, J.B., GRPANL: A Suite of Computer Programs for Analyzing Complex Ge and Alpha-Particle Detector Spectra, report UCRL-53861, Lawrence Livermore National Laboratory, Livermore, CA (1988).

[8] CLOSE, D.A., PRATT, J.C., MALANIFY, J.J., ATWATER, H.F., Nucl. Instrum. Methods Phys. Res. A **234** (1985) 556–561.

Table I

Comparison of measured results with declared values for several standards.

Standards	Measured % ^{235}U	% err.	Difference[a] %	σ	Standards	Measured % ^{235}U	% err.	Difference[a] %	σ
Small UO_2 powder	0.014	85	–23	0.3	UF_6 (0.055 kg)	3.442	1.3	–0.5	0.4
Depleted oxide	0.183	8.0	Unknown		Oxide (0.0064 kg)	4.061	1.8	–3.0	1.7
ECNRM171 (0.17 kg)	0.339	1.8	+5.9	0.7	(Same)	4.131	1.0	–1.4	1.4
Uranyl Nitrate	0.310	13	+1.9	0.1	(Same)	4.135	1.4	–1.3	0.9
Small UO_2 powder	0.488	3.6	+1.0	0.3	Oxide	4.520	0.7	+1.3	1.8
Metal (0.087 kg)	0.709	3.0	–0.1	0.0	ECNRM171 (0.2 kg)	4.551	0.6	+0.7	1.1
U_3O_8 (0.17 kg)	0.739	3.5	+4.0	1.1	Small UO_2 powder	4.949	1.1	+1.4	1.3
UO_2 powder (0.17 kg)	0.764	3.2	+7.6	2.4	Small UO_2 powder	10.09	0.6	+0.2	0.3
UO_2 pellets (0.05 kg)	0.721	3.8	+1.5	0.4	Oxide	17.40	0.6	+1.7	2.9
Small UO_2 powder	0.993	1.5	+0.2	0.1	Oxide	27.16	0.5	+0.2	0.4
Oxide (0.0064 kg)	1.925	3.2	+1.3	0.4	Small UO_2 powder	49.35	0.7	+0.0	0.0
(Same)	1.901	1.2	+0.0	0.0	Oxide	53.00	0.4	+1.7	4.2
(Same)	1.885	0.9	–0.8	0.9	Oxide	66.99	0.6	+1.3	2.1
ECNRM171 (0.2 kg)	1.967	1.4	–0.1	0.1	Small UO_2 powder	75.68	0.5	+0.7	1.4
Pellets (0.05 kg)	2.006	1.2	+0.8	0.6	Oxide	91.67	0.8	+0.6	0.8
Small UO_2 powder	1.984	1.6	–1.5	0.9	Oxide	92.54	1.1	–0.6	0.5
Pellets (0.05 kg)	2.846	1.1	–1.5	1.4	Metal disk (0.3 kg)	90.99	9.7	–2.3	0.2
ECNRM171 (0.2 kg)	3.026	1.0	+2.6	2.6	U_3O_8 (0.05 kg)	95.66	2.0	+2.3	0.6
Small UO_2 powder	2.993	1.0	-0.5	0.5	Oxide	92.79	2.9	–0.8	0.3
Oxide (0.0064 kg)	2.882	2.2	-4.8	2.2	Metal	96.14	3.8	+2.8	0.7
(Same)	3.042	0.9	+0.7	0.8	UF_6 (0.05 kg)	89.71	2.0	–4.2	2.1
(Same)	3.069	1.2	+1.6	0.8	Oxide	94.63	3.8	–2.5	0.7
(Same)	3.011	0.7	-0.3	0.5					

[a] Difference = measured – declared, expressed in % difference and in units of σ = (% difference)/(measured error).

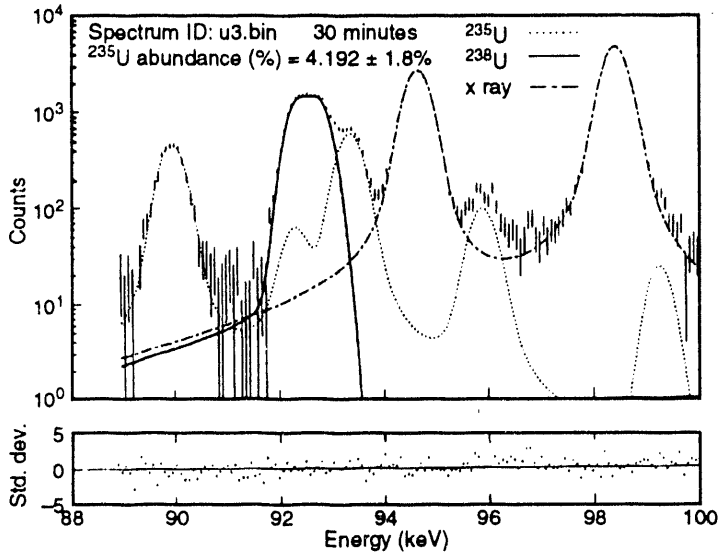

Fig. 1. This plot of the 88- to 100-keV region of a uranium spectrum shows the component responses for ^{235}U, ^{238}U, and uranium x rays. Each response is calculated from known nuclear parameters and from the energy-scaling and peak-width calibrations determined from other peaks in the spectrum. The enrichment calculation is based on measuring the relative intensities of the ^{235}U and ^{238}U responses.

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