URANIUM ISOTOPIC COMPOSITION ANALYSIS USING GAMMA SPECTROMETRY: APPLICATIONS IN NUCLEAR FORENSICS

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Abstract. This paper describes the methodology used for uranium isotopic composition analysis. It is shown how the computer software "Multigroup γ-ray Analysis Method for Uranium" (MGAU), together with gamma spectrometry, can be applied for categorization and characterization of the samples containing uranium. In addition, the specific recommendations for the detector choice are presented. This methodology is often used for nuclear forensic investigations, as the obtained results together with other types of information, may reveal the origin of the material. The tests results carried out with the use of certified reference uranium isotopic materials SRM 969 and CRM 146 showed a very good performance of the MGAU 4.2 code for an accurate characterization of ^{235}U and ^{234}U abundances. The experiment results were analyzed and some characteristics of the method were described. The real case study was taken as an example of application of this methodology. An alternative approach for categorization of samples containing uranium using Monte Carlo simulations was developed. An overview of this approach is presented, as well.

Key words: Gamma spectrometry, nuclear forensics, Uranium, MGAU.

1. INTRODUCTION

Following the collapse of the USSR, a large number of facilities using ionizing radiation were closed or destroyed. As a result, a large amount of radioactive sources, as well as nuclear material was lost, leading to the problem of orphan sources in the Black Sea region. In addition, the number of cases of illicit trafficking involving radioactive materials grew exponentially. This created the need to strengthen the capabilities of Eastern European countries in the new nuclear forensics field. The material of the most interest involved in the above mentioned cases is uranium, as one of its isotopes (U-235) can be used in the production of a nuclear bomb. While performing nuclear forensics analyses on a

seized sample which contains uranium, the first and main question which must be answered is, what is the isotopic composition of uranium present in the sample? There is a set of techniques capable of answering this question. The appropriate technique is selected with respect to the available equipment. Usually, it is important to preserve the samples for legal reasons. Therefore, the non-destructive assay (NDA) is preferred.

Gamma spectrometry is the most applicable, non-destructive technique used for uranium isotopic composition analysis. The detector has to be chosen appropriately for every specific type of measurement.

Upon characterizing nuclear material of unknown origin, some questions related to the production processes and intended end use may be addressed. If an appropriate knowledge base is also available, full attribution of materials to the point of loss-of-control might be possible [1].

1.1. DETECTOR CHOICE FOR THE MGAU

The MGAU [2, 3, 4, 5] code analyzes the complex region in uranium spectra centered around 100 keV which requires use of a high-resolution germanium detector in the acquisition of the spectrum.

The general recommendation for MGAU analysis is to have the detector energy calibration gain set at approximately 0.075 keV/channel. In order to obtain these gain settings, it is usually set 307 keV on 4096 channels. It is possible, however, to make measurements with different gain settings. Typically, for large Non-Destructive Assay (NDA) Systems, when a coaxial detector is used, the setting of 0.095 keV/channel gain is used to allow for an energy range up of to about 1.5 MeV in a single 16 k channels spectrum. MGAU can be used to analyze such spectra, and the code retrieves the initial energy calibration directly from the spectrum file or uses the user input [5].

2. PERFORMANCE TESTING OF THE URANIUM ISOTOPIC MULTI-GROUP ANALYSIS CODE MGAU 4.2 AND ANALYSIS OF URANIUM SPECTRA

2.1. MATERIALS AND METHODS

Gamma spectrometry measurements were performed on a Planar HPGe (High-Purity Germanium) Detector System with liquid nitrogen. The detector resolution (FWHM) at 122 keV is 575 keV. The Physical Characteristics of the detector are: Active Diameter – 36 mm, Active Area – 1000 mm^2, Thickness – 15 mm, Distance from Window (outside) – 14 mm, Window Thickness – 0.5 mm, Window Material – Al. The GL1015R detector is established in the Kiev Institute of Nuclear Research.

The energy calibration was performed automatically as the window in which the spectrum was collected was set to 4096 channels and approximately 307 keV.

Uranium isotopic standard reference materials SRM 969 and CRM 146, consisting of eight samples with different ^{235}U enrichments were used in the study. SRM 969 samples contained 200.1g of U_3O_8 powder (ρ = 2.5 g/cm^3) encased in Al cylindrical containers with internal dimensions Ø 70 × 21 mm. CRM 146 samples contained 230 g of U_3O_8 powder (ρ = 3.78 g/cm^3) placed inside similar Al containers with internal volume dimensions Ø 70 × 15.8 mm. Front wall thickness of the containers was 2 mm [3]. The certified isotopic abundances of the reference samples are shown in Table 1.

Table 1

Certified isotopic compositions of SRM 969 and CRM 146 standard reference material samples (wt%)

Material ID	$^{234}U/U$	$^{235}U/U$	$^{238}U/U$
NBS-031-116	0.0020 ± 0.0002	0.3206 ± 0.0002	99.6627 ± 0.0004
NBS-071-116	0.0053 ± 0.0002	0.7209 ± 0.0005	99.2738 ± 0.0004
NBS-194-116	0.0174 ± 0.0002	1.9664 ± 0.0014	98.0159 ± 0.0018
NBS-295-116	0.0284 ± 0.0004	2.9857 ± 0.0021	96.9826 ± 0.0029
NBS-446-116	0.0365 ± 0.0003	4.5168 ± 0.0032	95.4398 ± 0.0032
NBL0041	0.15076 ± 0.00037	20.311 ± 0.020	79.339 ± 0.020
NBL0042	0.3756 ± 0.0011	52.800 ± 0.042	46.560 ± 0.043
NBL0043	0.9849 ± 0.0029	93.233 ± 0.0053	5.4895 ± 0.0053

2.2. RESULTS

The standards SRM 969 and CRM 146 were measured. Eight spectra were collected with a planar HPGe detector GL1015R. The samples were placed at approximately 56 mm from the detector's cap. The live time was set to be 4000 seconds for every measurement. The geometry of every measurement was the same.

Figure 1 showed that the count rate in the peak with 185.71 keV energy, which corresponds to ^{235}U isotope is strictly linear with the enrichment. This rule is sometimes used for the determination of U enrichment by reproducing the same geometrical conditions.
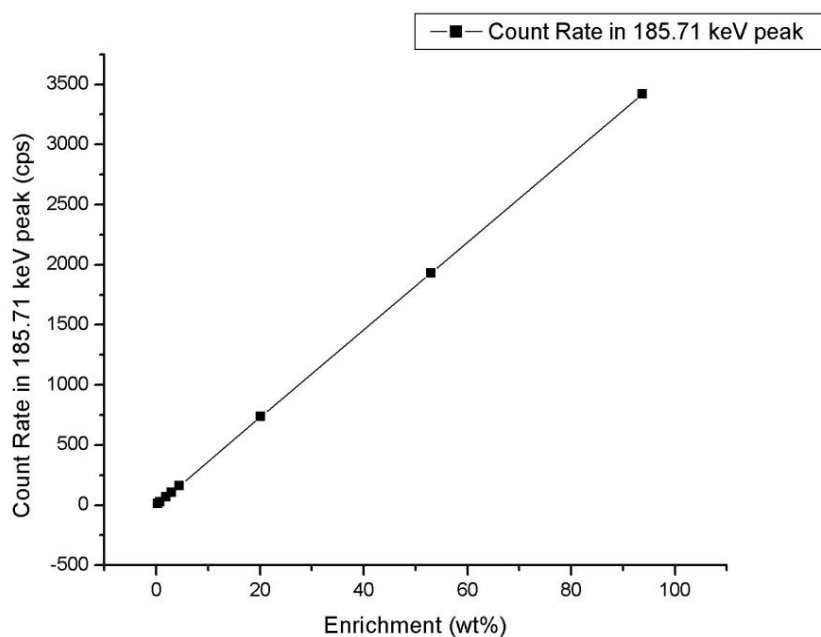
Fig. 1 – The count rate under 185.71 keV peak dependence of the enrichments of U.

Table 2

Measured isotopic compositions of SRM 969 and CRM 146 standard reference material samples
(wt%)

Material ID	^{234}U/U	^{235}U/U	^{238}U/U
NBS-031-116	0.0023 ± 0.0009	0.3206 ± 0.0182	99.6770 ± 0.0185
NBS-071-116	0.0053 ± 0.0010	0.7281 ± 0.0157	99.2666 ± 0.0159
NBS-194-116	0.0182 ± 0.0010	1.9653 ± 0.0187	98.0165 ± 0.0190
NBS-295-116	0.0290 ± 0.0011	2.9977 ± 0.0242	96.9733 ± 0.0245
NBS-446-116	0.0368 ± 0.0012	4.5064 ± 0.0326	95.4568 ± 0.0330
NBL0041	0.1482 ± 0.0022	20.1760 ± 0.1114	79.6758 ± 0.1128
NBL0042	0.3794 ± 0.0040	53.0107 ± 0.2187	46.6099 ± 0.2215
NBL0043	0.9778 ± 0.0106	93.7899 ± 0.6922	5.2323 ± 0.7

After comparing Tables 1 and 2, it is not difficult to observe that there are no significant differences in the results. However, the uncertainties in the experimental data are higher due to the fact that only gamma spectrometry was used to obtain this data, and not mass and alpha spectrometry as it is in the case of the certified material.

3. NUCLEAR FORENSICS INVESTIGATION: A CASE STUDY

3.1. MATERIALS AND METHODS

In August 2010, the Laboratory of Radiation Control (LRC) located in the Republic of Moldova received 1.8 kg of metal containing uranium which was seized in illicit trafficking. There were two items received, which were weighed and had their dimensions measured. The larger object (object 1) was frustoconical with a missing part. The inner and outer diameters were 34 mm and 85 mm, and the height 35 mm. The total weight of the object was 1760 g. The smaller object (object 2) was identified as the missing part of the object 1. Its mass was measured as 96.68 g (Fig. 2).

Fig. 2 – The objects seized in illicit trafficking.

3.2. GAMMA SPECTROMETRY SYSTEM

Gamma spectrometry measurements were performed on a broad energy Ortec HPGe detector which was not established in the environment with the low background. The detector resolution (FWHM) at 122 keV is 593 eV and at 1332 keV–1.7 keV. The energy calibration was performed with ^{152}Eu (122 keV and 344 keV) and ^{137}Cs (662 keV). After performing the energy calibration, we came up with approximately 0.097 keV/channel in the 0–300 keV region. This gain is used to allow for an energy range up to about 1.5 MeV in a single 16 k spectrum [7].

3.3. GAMMA SPECTROMETRY MEASUREMENT

The spectrum of the object 2 was collected for 4000 seconds (live time). The object was positioned 18 cm from the detector surface, which allowed for the dead time of 3 %.

By a careful analysis of the spectrum, most of the peaks were attributed to the photons originating from ^{235}U, ^{238}U and their daughters. No other radio nuclides could be detected under these experimental conditions, except the natural background.

The analysis shows that the sample does not contain detectable quantities of fission products or other artificial radio nuclides.

3.4. MGAU

The gamma spectrum of object 2 was considered suitable for MGAU analysis, due to the well-defined uranium peaks and absence of interfering radio nuclides. In addition, the object was in solid state and no container was used, which ensured that the attenuation of low energy gamma photons important for MGAU would be as low as possible. Finally, the sample thickness was adequate.

The MGAU analysis was never before performed in the Republic of Moldova. The limitations of the method were that the detector window was much thicker than recommended for MGAU, the environment was not low background and the detector was not of the recommended type (planar). Hence, in view of the mentioned limitations, in a first step, the code was tested on the natural uranium sample. The calculated composition (0.7 % of its weight) was in good agreement with the natural composition (0.711 %).

The results of the MGAU analysis are shown in Table 3. The results show that the analyzed object contains depleted uranium, with ^{235}U content of (0.364 ± 0.035) %.

Table 3

Results of the gamma spectrometric measurements and MGAU analysis

Sample Id.	Int. No.	^{234}U	unc.	^{235}U	unc.	^{238}U	unc.
DU2010	–	0.0013(%w)	0.0013	0.364(%w)	0.035	99.63(%w)	0.12

The method worked reasonably well and the results were similar to the ones included in the Institute of Transuranium Elements (ITU) report. The differences

were only observed in the uncertainties. This might be explained by the fact that in ITU, the planar HPGe detector with good resolution at low energy was used.

4. CATEGORIZATION OF SAMPLES CONTAINING URANIUM USING MONTE CARLO SIMULATIONS SOFTWARE GESPECOR AND HIGH RESOLUTION GAMMA SPECTROMETRY

Taking into account the fact that the method using MGAU requires large financial expenses and not all of the countries (especially Eastern Europe countries) are capable of upgrading their laboratories, we came with an idea to develop an alternative approach for uranium categorization. The overview of the developed and tested approach is described in the following paragraph.

The Canberra HPGe detector with liquid nitrogen was used. The window of energy spectrum was set to 8 k channels. The energy calibration was performed in order to identify the 185.71 keV peak which corresponds to ^{235}U isotope. This gamma line was used in the analyses. The detector resolution (FWHM) at 122 keV is 880 eV and at 1332 keV–1.82 keV.

4.1. RADIOACTIVE MATERIAL (RAM)

Gamma standard source containing ^{137}Cs was taken as a reference sample and a Standard Reference Material (SRM) 295 was taken as an unknown sample containing uranium.

4.2. EXPERIMENTAL SETUP

The container with uranium oxide was placed on the organic glass substrate at the 80.9 mm distance from the detector's cap (Fig. 3).

The certified point source of ^{137}Cs was placed in the center of organic glass, at the same distance from the detector as the container with U_3O_8.

4.3. GESPECOR

The Monte Carlo simulations software called GESPECOR [6, 8] was used for the determination of the full energy peak efficiencies of the 185.71 keV gamma line of ^{235}U and 661.65 keV gamma line of ^{137}Cs.

The geometry used in the experiment for the both radioactive materials, their characteristics, and the characteristics of the detector were transferred into GESPECOR (Fig. 3).
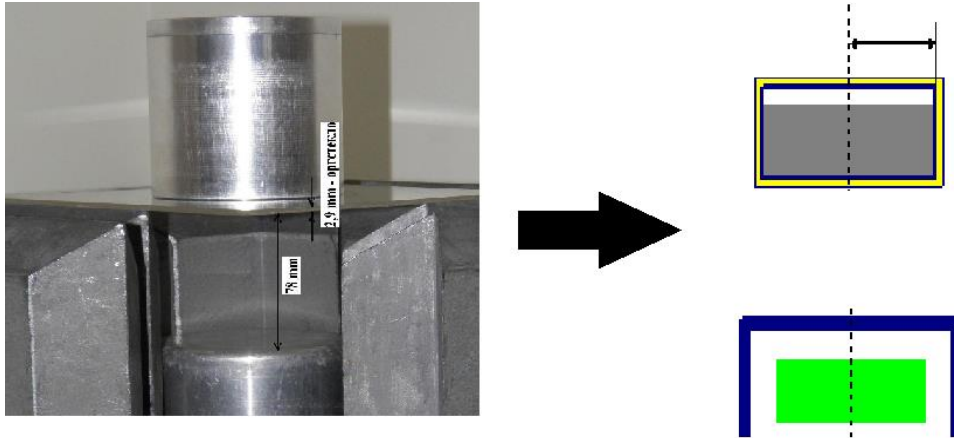
Fig. 3 – The geometry transfer to GESPECOR.

The data obtained from Monte Carlo simulations are presented in Table 4.

Table 4

The output of GEPECOR software

Nucl.	Decay	Energy	Yield	Fc	Nsec	Nsum	IdealEff.	Err
U-235	ALPHA	185.71	0.572	0.9994	2	0	0.231E-2	0.21

The correction factors "Fc" were insignificant for both cases and there was no need to take into account the true coincidence summing corrections for HPGe detector like it was done in [9]. Therefore, the full energy peak efficiencies (IdealEff., further referred to as ε) were taken directly from the output data of GESPECOR and were used in the calculations.

4.4. CALCULATIONS

A priori the calculations we used the same procedure for the ^{137}Cs reference point source in order to determine whether we transferred correctly the characteristics of the detector into GESPECOR software. The measured and the certified activities of the radioactive source where in good agreement with less than 5% difference. It was concluded that the detector was set correctly.

Further, we calculated the activity of [235]U in SRM-969-295 standard using (1)

$$A = \frac{I}{Y \cdot T \cdot \varepsilon},$$

(1)

where: A is experimental activity, I is the net area under the 185.71 keV peak, T is the time of measurement, and Y is emission probability of 185.71 keV peak, and ε is the full energy peak efficiency for the 185.71 keV peak.

$$A = \frac{10\,681\,684 \text{ counts}}{0.572 \cdot 20199.77 \text{ seconds} \cdot 0.023098} = 400 \text{ kBq}. \qquad (2)$$

Next, we converted the activity from (2) into the enrichment of uranium in the sample, and obtained 2.9571 (m%). Using the simulated full energy peak efficiency, we obtained that the analyzed sample contained 2.9571 (m%) enriched Uranium. We compared this result to the certified one, presented in Table 10 – 2.9492 (m%). It can be effortlessly observed that the experimental and certified results are in a very good agreement having a difference less than 0.5%.

The geometry of the experiment, chemical composition and density of the unknown sample has to be precisely determined. The reference material, like a point source, must be used to ensure a proper transfer of detector's characteristics to the Monte Carlo simulations software.

5. CONCLUSION

This paper describes most of the techniques applied for the uranium isotopic composition analysis using gamma spectrometry. The method using MGAU 4.2 was tested using certified reference material. It proved to give very good results with quite small uncertainties. The measurement results showed an excellent agreement with the declared data. It was demonstrated that MGAU version 4.2 can be successfully used for the uranium isotopic composition analyses and no specific knowledge in the field of nuclear physics and gamma spectrometry is required. The method is perfectly suited for the use not only in the laboratory, but also on-site and can be easily handled by non-physicists, as it requires no full energy peak efficiency calibrations. However, it had few limitations. The real case study was taken as an application example of this method.

The case study presented here is a typical case of nuclear material smuggling, seized in illicit trafficking in Eastern Europe. Performed analysis with available equipment was used for the laboratory's report presented in court.

The sample has proven to be a piece of depleted uranium (DU) which was stolen from an irradiating facility. It served as shielding for a strong gamma source, such as Iridium-192. Due to the fact that the fission products were not detected, it was concluded that the sample was made not out of spent fuel, but rather the leftover uranium from the enrichment facility. Both objects are categorized as orphan sources and are stored in the Radioactive Waste Repository of the Republic of Moldova.

In order to overcome the limitations of the MGAU described in [3], we came with the new approach for the determining of the enrichment of uranium. It is useful when no reference material is available or when the geometry of the sample which has to be analyzed does not correspond to the one of reference material, so the full energy peak efficiency calibration cannot be done by measurement. It is also useful when the MGAU code cannot be used due to the poor detector resolution. However, it required a good level of knowledge and skills in chemistry and Monte Carlo simulations. The main advantage of this method is that all the general applicability requirements as those of the MGAU can be ignored. However, it can mostly be applied for the determination of Uranium enrichment, as the 186.71 keV peak can be effortlessly distinguished due to its good emission probability. At the same time, this approach can be used for any other type of radioactive material, as long as it has the peak with no interferences and reasonably good emission probability.

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