Nondestructive Determination of $^{235}$U Enrichment of Uranium Samples in the Presence of Actinides ($^{232}$Th, $^{236}$U, $^{237}$Np)

Yu V Stogov

Department of Theoretical and Experimental Physics of Nuclear Reactors, National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe highway, 31, Moscow, Russia

YVStogov@mephi.ru

Abstract. The measurements were performed using a planar Ge detector. The multi-group analysis (MGA) for the determination isotopic abundances in low enriched uranium samples is applied in this study. In order to perform the analysis of complex gamma-spectrums, were taken a number of gamma-spectrums of single reference samples, containing $^{235}$U, $^{238}$U, $^{232}$Th, $^{236}$U, $^{237}$Np with declared isotopic abundance. The gamma-ray spectrums in the range of low energy X-ray and gamma-ray peaks of these samples were obtained and analyzed.

1. Introduction
The determination of $^{235}$U enrichment is of great importance of nuclear technology. The MGA for uranium (MGAU) method for non-destructive assay of $^{235}$U enrichment does not require the use of any nuclear material standards [1].

The MGAU-code is based on the analysis of the 89-120 keV region of Ge detector spectrum. There are some limitations of using of MGAU-code: the resolution at $E_\gamma=122$ keV of Germanium detector less than 700 eV; it requires from the daughter isotopes be in activity equilibrium with the $^{235}$U and $^{238}$U parent isotopes (it require up to six months after chemical separation); the accuracy of the method diminishes when the thickness of sample container becomes excessive; the abundances of actinides ($^{232}$Th, $^{236}$U, $^{237}$Np, $^{239}$Pu) and other gamma-emitter nuclides (fission products, et al) must be very little. The MGAU-code was designed to operate with little or no user interaction, that is as «complete secrecy»-code (we can not change the parameters of the code). Hence the conclusion: it is necessary to perform a preliminary analysis of the gamma-ray spectrums of the samples for the presence of «interfering» impurities - gamma emitters (actinides) [1-2].

The first task of this investigation is to perform the analysis of gamma-spectrums of single reference samples containing $^{235}$U, $^{238}$U, $^{232}$Th, $^{236}$U, $^{237}$Np with declared isotopic abundance.

The second task – using the MGAU-code to obtain the results of the $^{235}$U enrichment of samples: a) «pure» U-samples and b) «dirty» U-samples, containing different concentrations of actinides ($^{232}$Th, $^{236}$U, $^{237}$Np). Before each using of the MGAU-code it is necessary to analyze gamma-spectrum of sample.

The third task – to obtain the results of the $^{235}$U enrichment sample using the gamma-spectrum of investigated sample without use of MGAU-code. In order to decide this task the «peak-ratio» technique was applied [3] in the narrow 89-99-keV energy range. The peak 89.95-keV in spectrum of the sample is a measure of the $^{235}$U concentration (X-ray Th X $K_{a2}$). The intensities of 92.4 and 92.8-
keV lines of $^{234}$Th are used as a measure of $^{238}$U concentration of the sample. The 93.35-keV thorium Kα$_1$ line was used as a measure of $^{235}$U concentration. The contribution of 93.35-keV line was taken into account by using the special procedure.

2. Experimental description

2.1. Equipment
Spectrums were taken in planar Ge detector with resolution 46.0 eV at $E_\gamma=122$ keV (full width at half maximum, FWHM). The multichannel digital analyzer DSA-1000 Canberra and Genie-2000 (including MGAU-code) soft were used. The spectrums were stored in 4096 channels with a gain of 0.75 keV/channel. During the measurements there was performed the control of the position of the X-ray and gamma-ray peaks at energies of 63.1 and 185.72 keV, respectively belonging $^{234}$Th and $^{235}$U. The maxima of the peaks were situated respectively in the channels (844±1) and (2476±4).

2.2. The Gamma and X-rays Nuclear Data
The Gamma and X-rays nuclear data are presented in Table 1.

| Nuclide                  | Energy (keV) | Photon emission probability (%) |
|--------------------------|--------------|---------------------------------|
| $^{234}$U/$^{231}$Th    | 89.95±0.2    | 1.00±0.6                        |
| Th Kα$_2$                | 89.957       | 28.2±0.6                        |
| Pa Kα$_2$                | 92.282       | 28.3±0.6                        |
| $^{238}$U/$^{234}$Th     | 92.38        | 2.8±0.3                         |
| $^{235}$U/$^{231}$Th     | 92.80        | 2.8±0.3                         |
| Th Kα$_1$                | 93.02        | 0.047±0.006                     |
| U Kα$_3$                 | 93.844       | 0.098±0.003                     |

2.3. The Procedure of the Experimental Investigation

2.3.1. The Investigation of Influence of Different Concentrations of $^{232}$Th and $^{236}$U Presence in $\text{UO}_2$-Samples on the Results of $^{235}$U Enrichment Determination by MGAU-code

The $^{235}$U enrichment of the reference $\text{UO}_2$-sample ($^{235}$U enrichment x=6.50 wt%) was determined by MGAU-code (Table 2).

| Nuclide | Weight fraction of nuclide, (%) | Derivation |
|---------|---------------------------------|------------|
| $^{234}$U | 0.045                          | 0.010 (22.2 %) |
| $^{235}$U | 6.352                          | 0.084 (1.3%)  |
| $^{238}$U | 93.603                         | 0.084 (0.1%)  |
The gamma-spectrum of the reference UO₂-sample (²³⁵U enrichment x=6.50 wt%) was taken. The peak 89.95-keV in spectrum of this sample is a measure of the ²³⁵U concentration (X-ray Th X Kα₂) (Figure 1).

The gamma-spectrum of ²³²Th sample was taken. In this spectrum the peak of sample 89.95-keV is a measure of ²³²Th concentration.

The count rate in peaks 89.95 keV in both samples was determined (measuring peak intensity in counts/s). The time of measuring ²³²Th sample spectrum, corresponding the increasing the value of count rate in peak 89.95 keV in the spectrum of UO₂-sample (²³⁵U enrichment x=6.50 wt%) is obtained.

Using the gamma-spectrum of UO₂-sample contained in the memory of multi-channel analyzer, the UO₂-sample was removed from Ge - detector and then the ²³²Th-sample was put on the detector. After the spectrum measurement of ²³²Th-sample during the necessary time the count rate in the peak 89.95 keV increased by 1%. That corresponds to an increase of weight fraction ²³²Th in the mixture of (UO₂+²³²Th). A new treatment of obtained spectrum was performed by MGAU-code, ect. Step by step we obtain the response function of MGAU-code of increasing 89.95 keV count rate peak by gamma-emitter (²³²Th-sample). This count rate depends from parameter P:

\[
P = \frac{S_{90\text{ keV}}(UO_2(6.5\%)) + 232\text{Th}}{S_{90\text{ keV}}(UO_2(6.5\%))}
\]

The dependence of ²³⁵U enrichment (obtained using MGAU-code analysis) from parameter P is presented on Figure 2. The number of the «big points» on the Figure 2 corresponds that there are no message from MGAU-code «gamma-emitting nuclides are present in investigated sample» on measure of increase of parameter P before certain moment. The analogical procedure was performed with ²³⁶U sample. The results are presented in Figure 3 in dependence of the parameter \[P_1 = \frac{S_{90\text{ keV}}(UO_2(6.5\%)) + 236\text{U}}{S_{90\text{ keV}}(UO_2(6.5\%))}\].
**Figure 2.** Dependence of $^{235}\text{U}$ enrichment (obtained using MGAU-code analysis) from parameter $P$: $^{232}\text{Th}$ gamma-emitting nuclide is present in investigated sample.

**Figure 3.** Dependence of $^{235}\text{U}$ enrichment (obtained using MGAU-code analysis) from parameter $P_1$: $^{236}\text{U}$ gamma-emitting nuclide is present in investigated sample.
2.2.2. The «Peak-Ratio» Technique for $^{235}$U Enrichment Determination in Samples (Without Using of MGAU-Code)

On the Figure 4 is presented the gamma and X-ray spectrum of depleted uranium sample ($x=0.0025$ wt%). On the Figure 5 is presented the gamma and X-ray spectrum of enriched uranium sample ($x=98$ wt%)

![Gamma and X-ray spectrum of depleted uranium sample (x=0.0025 wt%).](image)

![Gamma and X-ray spectrum of enriched uranium sample (x=98 wt%).](image)

Figure 4. Gamma and X-ray spectrum of depleted uranium sample ($x=0.0025$ wt%).

Figure 5. Gamma and X-ray spectrum of enriched uranium sample ($x=98$ wt%).
The «strip» function of the multichannel analyzer was used as follows: 1) store an «enriched» spectrum of uranium enriched to 99.5% in $^{235}$U; 2) obtain the sample spectrum, then normalize the «enriched» spectrum to the 89.96 keV X-ray of the sample spectrum; and 3) subtract the normalized «enriched» spectrum from the sample spectrum. The resultant stripped spectrum contains the cleanly resolved $^{238}$U gamma-ray doublet at 92-keV peak from the stripped spectrum and the 89.96-keV X-ray peak area from the original sample spectrum are used to obtain the $^{238}$U/$^{235}$U ratio.

A relationship between enrichment, $X$, and the $^{238}$U/$^{235}$U ratio, $R$, can be obtained as follows:

$$X = \frac{100 \cdot w.f.^{235}U}{w.f.^{234}U + w.f.^{235}U + w.f.^{236}U + w.f.^{238}U}$$

(2),

where $w.f.^{234}U$, $w.f.^{235}U$, $w.f.^{236}U$, $w.f.^{238}U$ - weight fraction of $^{234}U$, $^{235}U$, $^{236}U$, $^{238}U$, respectively.

Substitutive the following approximation into the above equation:

$$w.f.^{234}U + w.f.^{236}U = 0.01 \cdot w.f.^{235}U$$

(3).

Than rearrange the terms to obtain:

$$X = \frac{100}{1.01 + \frac{w.f.^{238}U}{w.f.^{235}U}} = \frac{100}{1.01 + K \cdot R},$$

where $R = \frac{S_{92\text{keV}}}{S_{90\text{keV}}}$; $K = 5.46$ is an experimental constant. The constant $K$ is determined in by iterative way to give the best fit to the calibration data where $R = \frac{S_{92\text{keV}}}{S_{90\text{keV}}}$. The relationship between peak area ratios $S_{92\text{keV}}/S_{90\text{keV}}$ and $^{235}$U enrichment are presented in figure 6.

![Figure 6](https://example.com/figure6.png)
3. Conclusions
Using the high resolution planar Ge detector it is possible to take the gamma-spectrums of «clean» uranium samples and «unknown» «dirty» uranium samples, containing some concentrations of actinides. Using a single reference samples, containing $^\text{232}\text{Th}$, $^\text{235}\text{U}$, $^\text{237}\text{Np}$ etc. with declared isotopic abundance, it is possible to increase the count rates in corresponding gamma-spectrum peaks of «unknown» «dirty» uranium samples and obtain the response function of MGAU-code from the $^\text{235}\text{U}$ enrichment (using MGAU-code) in dependence of parameter $P$. At last, now it is possible to estimate the number of steps, that corresponds the increasing of $P$ on 1%, when the MGAU-code message «gamma-emitters are present in sample» will be obtain.

The «peak-ratio» technique description, presented in this paper, allows explaining the procedure of $^\text{235}\text{U}$ enrichment determination of uranium samples without using of MGAU-code and its allows obtaining the $^\text{235}\text{U}$ enrichment of investigated samples.

The study of nondestructive determination of $^\text{235}\text{U}$ enrichment of uranium samples in the presence of actinides will be continued.

4. References
[1] Gunnik R et al 1994 A New Analysis Code for Measuring U-235 Enrichments in Arbitrary Samples IAEA Symposium on International Safeguards, Vienna, Austria. Lawrence Livermore National Laboratory. Report UCRL-JC-114713 Livermore, California.
[2] Yucel H 2007 The Applicability of MGA Method for Depleted and Natural Uranium Isotopic Analysis in the Presence of Actinides ($^\text{232}\text{Th}$, $^\text{237}\text{Np}$, $^\text{233}\text{Pa}$ and $^\text{241}\text{Am}$) Applied Radiation and Isotopes 65 p 1269-1280.
[3] Passive Nondestructive Assay of Nuclear Materials 1991 NUREG/CR-5550 LA-UR-90-732 Edited by: Reilly D, Ensslin N and Smith H.
[4] Firestone R B, Shirley V S 1996 Table of Isotopes. Wiley, New-York.