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 Eγ=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 all) 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 $_{K\alpha 2}$). 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 $K\alpha_1$ 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 460 eV at E_γ=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.

Table 1. Gamma and X-rays nuclear data [4].

	Energy (keV)	Photon emission probability (%)
γ-ray(^{235}U/^{231}Th)	89.95±0.2	1.00±0.6
Th $_{K\alpha2}$	89.957	28.2±0.6
Pa $_{K\alpha2}$	92.282	28.3±0.6
γ-ray (^{238}U/^{234}Th)	92.38	2.8±0.3
γ-ray (^{238}U/^{234}Th)	92.80	2.8±0.3
γ-ray (^{235}U/^{231}Th)	93.02	0.047±0.006
Th $_{K\alpha1}$	93.35	46.0±0.9
U $_{K\alpha3}$	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 UO_2-Samples on the Results of ^{235}U Enrichment Determination by MGAU-code

The ^{235}U enrichment of the reference UO_2-sample (^{235}U enrichment x=6.50 wt%) was determined by MGAU-code (Table 2).

Table 2. The results of ^{235}U enrichment using MGAU-code analysis for UO_2-sample (x=6.50 wt%).

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 UO_2-sample (^{235}U enrichment x=6.50 wt%) was taken. The peak 89.95-keV in spectrum of this sample is a measure of the ^{235}U concentration (X-ray Th X $_{K\alpha 2}$) (Figure 1).

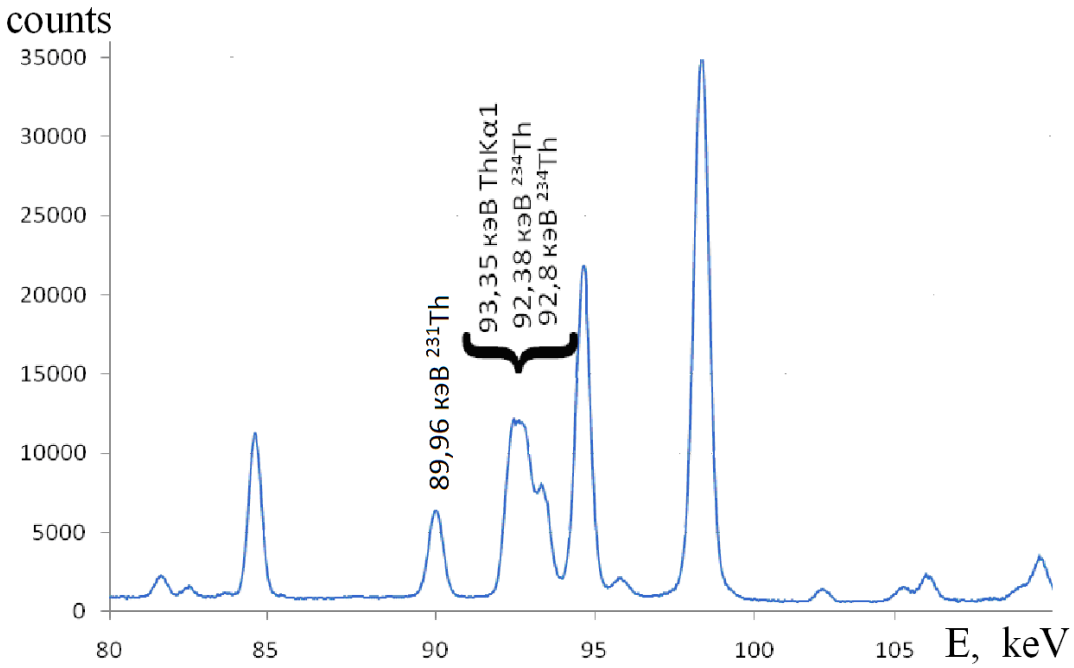

Figure 1. Gamma and X-ray spectrum of enriched uranium sample (x=6.50 wt%).

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

The count rate in peaks 89.95 keV in both samples was determined (measuring peak intensity in counts/s). The time of measuring ^{232}Th sample spectrum, corresponding the increasing the value of count rate in peak 89.95 keV in the spectrum of UO_2-sample (^{235}U enrichment x=6.50 wt%) is obtained.

Using the gamma-spectrum of UO_2-sample contained in the memory of multi-channel analyzer, the UO_2-sample was removed from Ge - detector and then the ^{232}Th-sample was put on the detector. After the spectrum measurement of ^{232}Th-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 ^{232}Th in the mixture of (UO_2+^{232}Th). 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 (^{232}Th-sample). This count rate depends from parameter P:

$$P = \frac{S^{90\,keV}(UO_2(6.5\%) + {}^{232}Th)}{S^{90\,keV}(UO_2(6.5\%))} \qquad (1)$$

The dependence of ^{235}U 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 ^{236}U sample. The results are presented in Figure 3 in dependence of the parameter $P_1 = \frac{S^{90\,keV}(UO_2(6.5\%) + {}^{236}U)}{S^{90\,keV}(UO_2(6.5\%))}$.

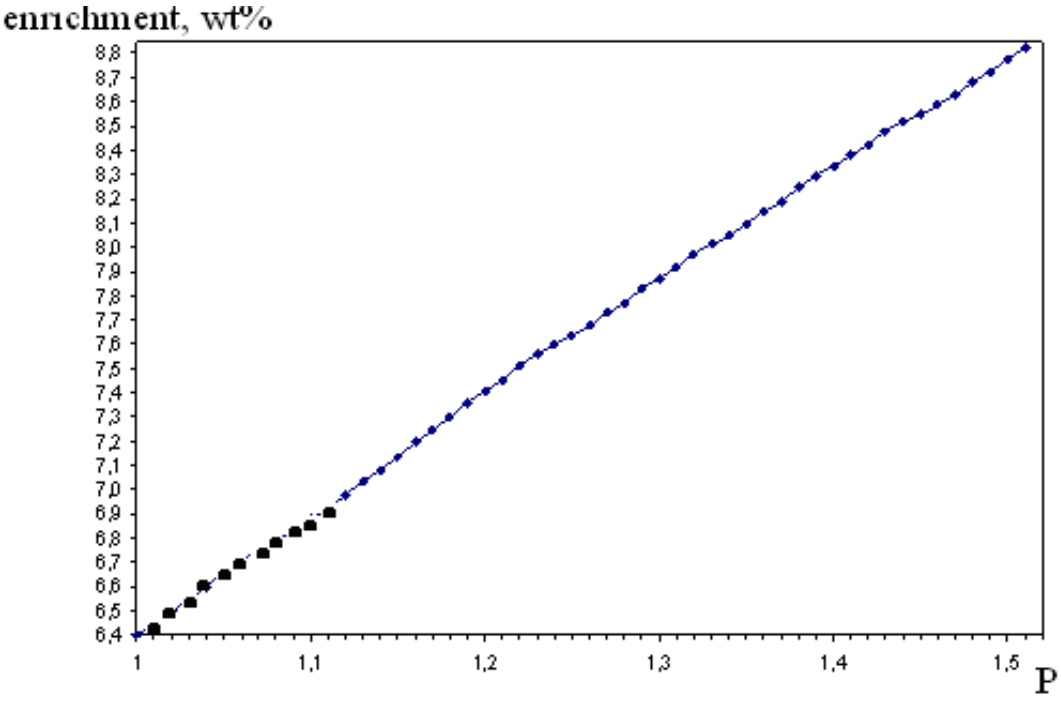

Figure 2. Dependence of ^{235}U enrichment (obtained using MGAU-code analysis) from parameter P: ^{232}Th gamma-emitting nuclide is present in investigated sample.

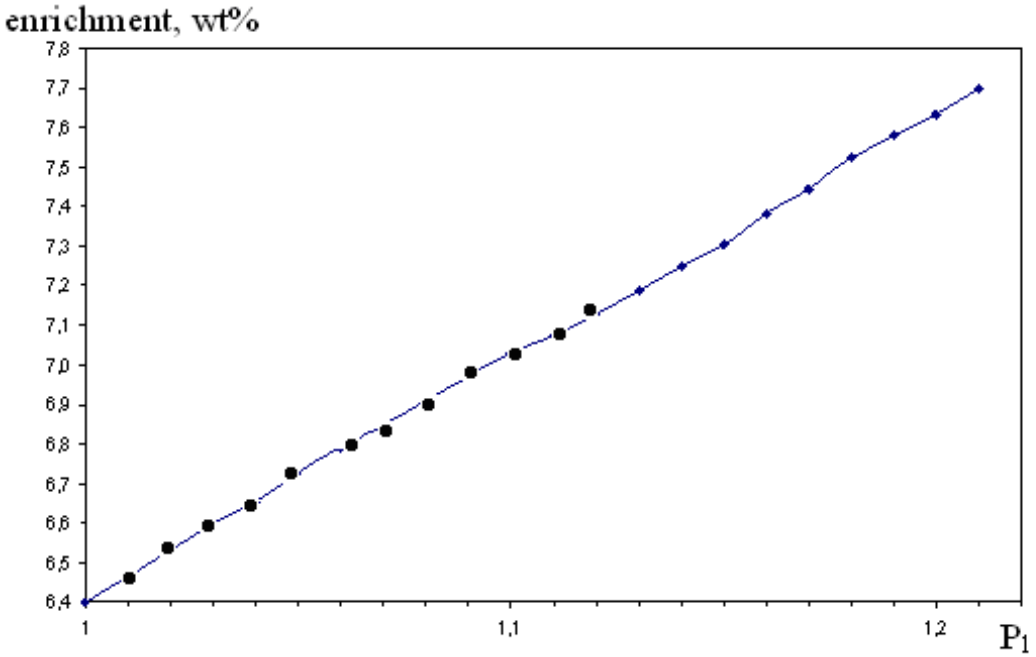

Figure 3. Dependence of ^{235}U enrichment (obtained using MGAU-code analysis) from parameter P_1: ^{236}U gamma-emitting nuclide is present in investigated sample.

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%)

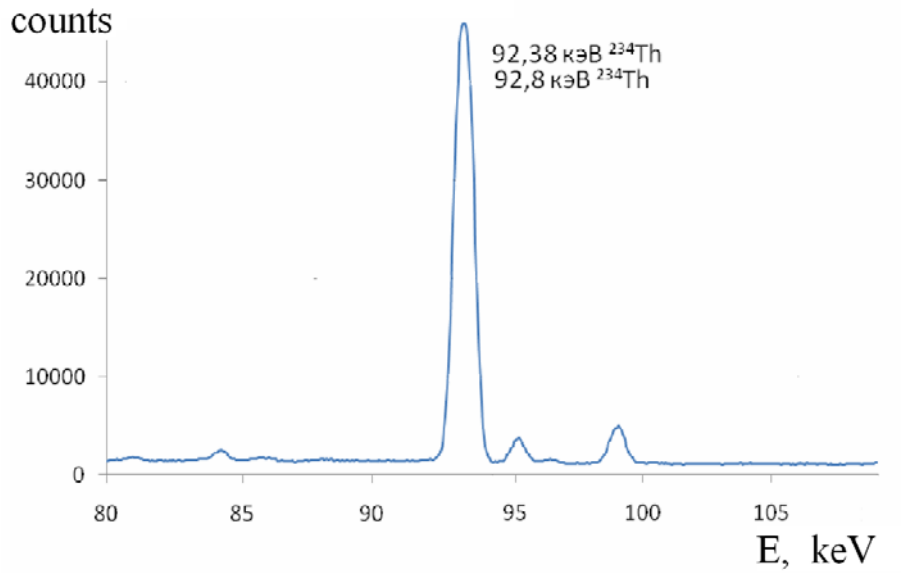

Figure 4. Gamma and X-ray spectrum of depleted uranium sample (x=0.0025 wt%).

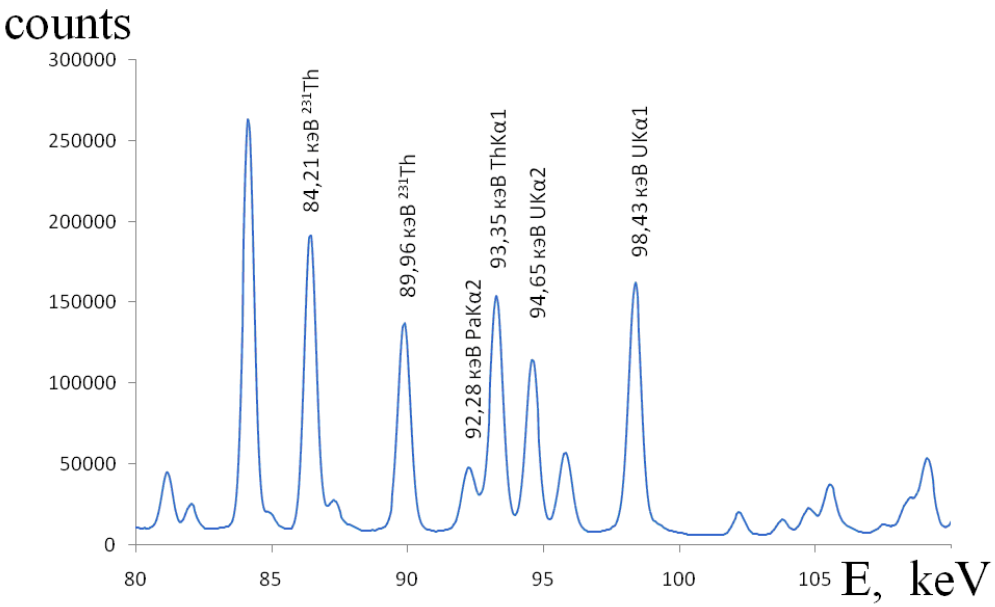

Figure 5. Gamma and X-ray spectrum of enriched uranium sample (x=98 wt%).

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:

$$X = \frac{100 \cdot w.f.^{235}U}{w.f.^{234}U + w.f.^{235}U + w.f.^{236}U + w.f.^{238}U} \quad (2),$$

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:

$$w.f.^{234}U + w.f.^{236}U = 0.01 \cdot w.f.^{235}U \quad (3).$$

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 $R = \frac{S^{92\,keV}}{S^{90\,keV}}$; $K = 5{,}46$ is an experimental constant. The constant K is determined in by iterative way to give the best fit to the calibration data where $R = \frac{S^{92\,keV}}{S^{90\,keV}}$. The relationship between peak area ratios S^{92keV}/S^{90keV} and ^{235}U enrichment are presented in figure 6.

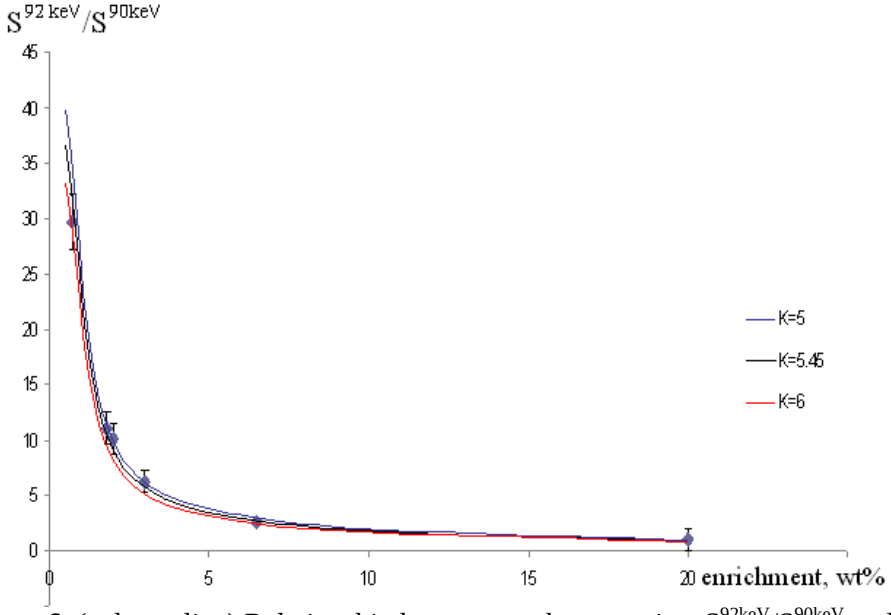

Figure 6. (color online) Relationship between peak area ratios S^{92keV}/S^{90keV} and ^{235}U enrichment.

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 ^{232}Th, ^{236}U, ^{237}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 ^{235}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 ^{235}U enrichment determination of uranium samples without using of MGAU-code and its allows obtaining the ^{235}U enrichment of investigated samples.

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

4. References

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