

## EXPERIMENTAL DETERMINATION OF THE URANIUM ENRICHMENT RATIO\*

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During the last years, as the international illicit traffic of radioactive / fissionable materials have increased, it became important to be able to apply fast and reliable methods for the uranium enrichment determination. This work describes such a method based on high resolution gamma- and X-rays spectrometry, using high purity Germanium (HP Ge) semiconductor detectors. The main problem of the method is the determination of the  $^{238}\text{U}$  activity, as this radionuclide is an alpha-rays emitter (its gamma-ray emissions are practically negligible). The solution found was to analyze the  $\text{K}_\beta\text{X}$ -rays region: (111...115) keV. High quality energy and efficiency detector calibrations are needed, too. The method was applied for several samples of small volume (powders of uranium oxide, enclosed in cylindrical plastic capsules). As a test of this method, an enriched uranium standard was measured; the obtained value was in agreement, within the estimated uncertainty, to the enrichment value stated by the producer of the standard (10%).

*Key words:* uranium, enrichment, gamma-rays, X-rays, spectrometry, activity, spectrum.

### 1. INTRODUCTION

Uranium is probably the most important radioactive element present in the nature. Most of the heat generating from the Earth's core is produced by the decay of the elements from the  $^{238}\text{U}$  radioactive series; this heat allowed the development and evolution of the first life forms on our planet. Nowadays, uranium is often used in nuclear power plants to produce electricity. Unfortunately, another application of uranium is the production of the most destructive weapons ever used in wars: the atomic bombs.

The natural uranium is a mixture of three radioactive isotopes [1]:  $^{238}\text{U}$  (99.2742% abundance),  $^{235}\text{U}$  (0.7204% abundance) and  $^{234}\text{U}$  (0.0054% abundance).

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Most of the applications of uranium are based on the energy generated by the fission of the  $^{235}\text{U}$  nuclide; so, it is important to increase its abundance in the uranium chemical compound used (to obtain enriched uranium). Sometimes, on the contrary, the abundance of  $^{235}\text{U}$  is reduced (to obtain depleted uranium), to produce containers for radioactive sources, ammunition etc.. The enriched uranium can be [2]: “highly-enriched uranium” (more than 20% of  $^{235}\text{U}$ ) and “low-enriched uranium” (less than 20% of  $^{235}\text{U}$ ) – with the variant “slightly-enriched uranium” (between 0.9% and 2% of  $^{235}\text{U}$ ). The enriched uranium is usually produced by using the diffusion (thermal or gaseous) and gas centrifuge methods.

The determination of the uranium enrichment is very important in various fields [3]: nuclear energetics, nuclear safeguards, radiation protection and especially, fight against illicit international traffic of radioactive materials and nuclear terrorism.

This paper presents a method for determining the uranium enrichment of small volume samples.

## 2. METHOD AND EXPERIMENTAL SET-UP

The uranium enrichment,  $e$ , expressed in %, is defined as:

$$e = \frac{m(235)}{m(235) + m(238)} \cdot 100, \quad (1)$$

where  $m(235)$  and  $m(238)$  are the masses of  $^{235}\text{U}$  and respectively  $^{238}\text{U}$ ; generally, the mass of  $^{234}\text{U}$  is very small compared to the masses of the two other uranium isotopes and it was neglected in this formula.

Considering the fact that both radionuclides are radioactive, the enrichment can be expressed as a function of the activity values of  $^{238}\text{U}$  and  $^{235}\text{U}$ . Starting from the basic formula:

$$\Lambda = \frac{\ln 2}{T_{1/2}} \cdot N, \quad (2)$$

where  $\Lambda$  is the activity,  $N$ -the number of radioactive nuclei and  $T_{1/2}$  – the half-life, the masses of the two isotopes are:

$$m(235) = \frac{\Lambda(235) \cdot 235 \cdot T_{1/2}(235)}{N_A \cdot \ln 2}, \quad (3)$$

$$m(238) = \frac{\Lambda(238) \cdot 238 \cdot T_{1/2}(238)}{N_A \cdot \ln 2}, \quad (4)$$

where  $\Lambda(235)$  and  $\Lambda(238)$  are the activity values of  $^{235}\text{U}$  and respectively  $^{238}\text{U}$ ;  $T_{1/2}(235) = 2.5706 \cdot 10^{11}$  years and  $T_{1/2}(238) = 1.633 \cdot 10^{12}$  years are their half-lives and  $N_A$  is the Avogadro's constant.

Using the formulae (1), (3) and (4), the enrichment of a sample containing uranium can be computed as:

$$e = \frac{1}{1 + \frac{\Lambda(238)}{\Lambda(235)} \cdot \frac{T_{1/2}(238)}{T_{1/2}(235)} \cdot \frac{238}{235}} \cdot 100 \quad (5)$$

The  $^{235}\text{U}$  nuclide emits both alpha and gamma-rays; the main photon emissions have an energy of 185.7 keV (0.572 emission intensity).  $^{238}\text{U}$  is practically a pure alpha emitter, with negligible gamma-ray emissions; however, this nuclide have several important X-ray emissions:  $\text{K}_{\alpha 2}$  (94.65 keV, with 0.157 intensity),  $\text{K}_{\alpha 1}$  (98.43 keV, with 0.253 intensity),  $\text{K}_{\beta 1}$  (111 keV, with 0.092 intensity) and  $\text{K}_{\beta 2}$  (114.9 keV, with 0.0311 intensity).

These properties recommend the use of the high resolution gamma-ray spectrometry method, in order to measure the activity values of  $^{238}\text{U}$  and  $^{235}\text{U}$ , and determine the enrichment of each sample. The activity values were computed according to the formula:

$$\Lambda = \frac{A}{t \cdot s \cdot \varepsilon} \quad (6)$$

where  $A$  is the net area of the peak corresponding to a gamma or X-rays emission of the nuclide,  $t$  is the measurement time,  $s$  is the photon emission intensity and  $\varepsilon$  is the detection efficiency of the full-energy absorption peak considered. The two peaks chosen to be measured in the spectrum are associated to the 185.7 keV gamma-rays ( $^{235}\text{U}$ ) and 111 keV X-rays ( $^{238}\text{U}$ ).

The measurement equipment used is composed of: HP Ge semiconductor detector (25% relative efficiency and 2.0 keV resolution at 1332.5 keV), preamplifier, spectroscopy amplifier, high voltage power supply, Multichannel analyzer (MCA) with 4 096 channels, Accuspec software package v. 7.3c (from CANBERRA Inc., USA). This spectrometer was energy and efficiency calibrated, by using point standard sources ( $^{241}\text{Am}$ ,  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$ ), produced by the Radionuclide Metrology Laboratory from IFIN-HH. The experimental data of the efficiency versus energy were fitted by the EFFIGIE computer code [4], while the deconvolution of the  $^{238}\text{U}$  KX-rays spectral region was performed with the COLEGRAM software [5].

The analyzed samples (eleven), with an unknown enrichment, consisted of solid (powder) uranium oxide –  $\text{UO}_2$ , closed in plastic capsules (25 mm diameter and 10 mm high). At the beginning, an enriched uranium standard, with similar characteristics was measured, and the result compared with the stated enrichment value (10%).

Afterwards, the standard was used to determine experimentally the enrichment of the all the samples. All the measurements were performed with the samples placed directly on top the detector, centered.

### 3. RESULTS AND DISCUSSION

The gamma-rays spectrum (including the KX-rays region of  $^{238}\text{U}$ ) of a sample containing enriched uranium is presented in Fig. 1; in the spectrum, besides the contributions of the uranium isotopes, the gamma-ray emissions of  $^{234}\text{Th}$  (63.3 keV) and  $^{234\text{m}}\text{Pa}$  (1 001 keV) – both nuclides from the  $^{238}\text{U}$  radioactive series, can be observed, too.

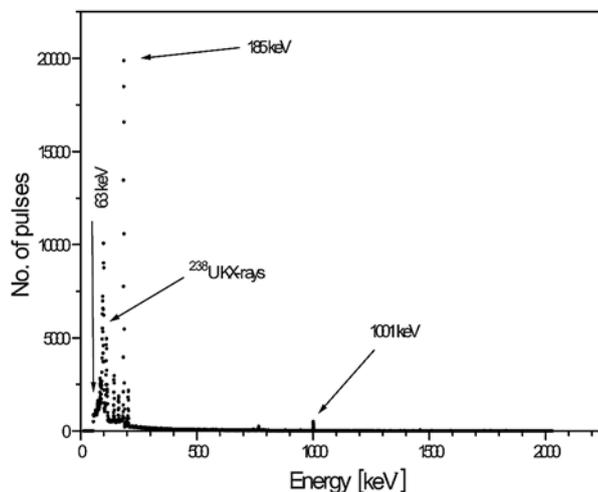


Fig. 1 – The gamma-rays spectrum of an enriched uranium sample ( $\text{UO}_2$ ).

The measurement of the uranium standard enrichment (code UMETA-1), according to the formulae presented above, gave a result of 8.8%; as the absolute uncertainty ( $k = 1$ ) of the enrichment value was 0.6%, the obtained result is in agreement with the value stated for the standard (10% with an absolute uncertainty of about 0.5%). The uncertainty components are due to: counting statistics, efficiency calibration (the region of the X-rays corresponds to the maximum of the efficiency versus energy curve), intensity of the gamma/X-rays emission, deconvolution of the  $^{238}\text{U}$  KX-rays spectral region and (small) geometrical differences between the activity standards used for calibration and the standards.

For the uranium oxide samples, the detection efficiency at 111 keV was calculated considering the enrichment standard, in order to minimize some of the problems mentioned above. The obtained results are presented in Table 1.

The relative uncertainties of the  $^{235}\text{U}$  activity values reported in Table 1 are usually between 3% and 4%, while for  $^{238}\text{U}$ , they are about 12%.

Future improvements of this method require a more accurate analysis of the uranium KX-rays spectral region and the use of several standards. Other methods, based on relative efficiency calibration should be used, too; their advantage is the possible analysis of volume samples, as the photons absorption is taken into account.

Table 1

Uranium enrichment for UO<sub>2</sub> samples

No.	Sample's code	$\Lambda(235)$ , Bq	$\Lambda(238)$ , Bq	$e$ , %
1	IFTAR 11	1 089	2 143	7.3
2	IFTAR 13	1 598	1 978	11
3	IFTAR 14	17 126	10 065	21
4	IFTAR 16	4 306	4 251	14
5	IFTAR 17	277	128	25
6	IFTAR 19	3 668	2 324	20
7	IFTAR 20	527	698	11
8	IFTAR 27	430	1 483	4.3
9	IFTAR 36	703	600	15
10	IFTAR 41	294	453	9.2
11	IFTAR 63	178	197	12

#### 4. CONCLUSIONS

- The use of high-resolution gamma-ray spectrometry allowed the determination of the enrichment for some small volume uranium oxide samples.
- The activity of <sup>238</sup>U can be determined by measuring its KX-rays, but special software for deconvolution of spectra must be used.
- The method was tested by using an uranium enrichment standard (10%).
- Future improvements are needed in order to measure volume samples and obtain higher accuracy results.

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#### REFERENCES

1. J. K. Tuli, *Nuclear Wallet Cards*, Brookhaven National Laboratory, Upton, New York, U.S.A., 2005, 70.
2. [http://en.wikipedia.org/wiki/Uranium\\_enrichment](http://en.wikipedia.org/wiki/Uranium_enrichment) (web page on the Internet).
3. J. Morel, C. Hill, M. Bickel, A. Alonso-Munoz, S. Napier, B. Thauvel, European Safeguards Research and Development Association NDA-WG Members, *Results from the international evaluation exercise for uranium enrichment measurements*, Applied Radiation and Isotopes, vol. 52, 509–522, 2000.
4. J. Morel, *Determination de la reponse en efficacite des detecteurs gamma-X par lissage de points experimentaux: code EFFIGIE*, LPRI/CEA, Saclay, France, 1996.
5. H. Ruellan, M. C. Lepy, J. Plagnard, *Presentation du logiciel "COLEGRAM"*, Note Technique LPRI/95/016, CEA, Saclay, France, 1995.