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CALIBRATING A LOW GAMMA COUNTING SYSTEM FOR MEASURING DEPLETED AND LOW ENRICHED URANIUM

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Abstract – The need for determining the $^{235}\text{U}/^{238}\text{U}$ ratio in environmental and biological samples is increasing, mostly after the Gulf War and the conflict on the Balkans. This paper describes a simple way to calibrate a low gamma counting system for measuring depleted, natural, and low enriched uranium.

Keywords: depleted uranium, calibration, uranium isotopic ratio

1. INTRODUCTION

The use of depleted uranium (DU) by the military in the Gulf War, and in the conflict on the Balkans has caused considerable concern by the general public. NATO-Member States reported incidences of health effects and lead to speculations on their potential link to an increased exposure by soldiers to DU in the line of duty [1]. Moreover, claims have been made about the possible threat to the health of residents in DU-affected areas. The United Nations Environment Programme (UNEP) sponsored DU post-conflict environmental assessment in Kosovo, Serbia, and Montenegro [2,3]. During the first surveys made by UNEP in Kosovo a NaI(Tl) detector with an alarm signal capable to be heard above the traffic noise and the wind was used [2]. These surveys were carried out to locate potential DU contaminated sites. Ge(Li) detectors were not used because it was not possible to transport the liquid nitrogen that was essential to cool them before applying bias voltage. However, after the publication of the surveys in Kosovo, the Yugoslavian authorities invited a UNEP team to carry out DU studies in Serbia and Montenegro [3]. This time a large number of soil samples were collected to be measured later on. The natural uranium levels of the soils at the site of collection range from 1.0 to 9.5 mg U.kg⁻¹ soil [3]. Taking into account the 0.7% ^{235}U abundance in natural uranium, DU was defined by the UNEP team as having a ^{235}U abundance in soil samples equal to, or less than 0.35% [3]. From the soil samples collected in Serbia and Montenegro it was determined that approximately 42% of these samples presented more than 10% DU. Here one must bear in mind

that farmers working in DU contaminated soils, as well children playing in such soils might sometimes ingest small amounts of DU [4-8]

One should be well prepared to evaluate the potential health and environmental impacts associated with the possibility of future military use of DU.

As a matter of fact, the issue of DU led several investigators to review recently the gamma spectrometric techniques to discriminate between natural uranium (NU) extant in the environment and DU in contaminated areas [3-8].

This paper proposes a simple method to analyze gamma spectra in order not only to discriminate DU and low enriched uranium (LEW) from NU, but also to establish the degree of uranium depletion or low enrichment. To do so, advantage was taken from a low gamma counting system, described elsewhere, which was put together to measure naturally occurring radioactive material (NORM), and technologically enhanced NORM (TENORM) [9].

2. FIRST DU GAMMA SPECTROMETRY

In the years seventies, when the use of lithium-drifted germanium detector [Ge(Li)] was still a novelty, few investigators dared to attempt to measure the ^{238}U through the 63.3 keV transition line from the daughter ^{234}Th . This was so, until isotopic ratios $^{235}\text{U}/^{238}\text{U}$ (reported originally as $^{238}\text{U}/^{235}\text{U}$) were successfully determined by gamma spectrometry in soils with NU content, and contaminated with DU, and then compared with the isotopic ratios obtained by mass spectrometry [10]. Soils with NU content were obtained at the Livermore California Valley and at Yosemite National Park, while those soils with DU contamination were collected at a high explosive test area [10]. TABLE I summarizes the isotopic ratios $^{235}\text{U}/^{238}\text{U}$ in natural and contaminated soils estimated from the data published in reference [10].

TABLE I. Isotopic ratio ($^{235}\text{U}/^{238}\text{U}$) in soils with NU and contaminated with DU as measured by gamma- and mass spectrometries. Based on data taken from ref. [10].

| Soil type | $^{235}\text{U}/^{238}\text{U}$ | |
|--|---------------------------------|-------------------|
| | γ spectrometry | mass spectrometry |
| Low uranium level with "natural" isotopic ratio | 0.027 ± 0.017 | 0.045 ± 0.001 |
| | 0.036 ± 0.016 | 0.047 ± 0.001 |
| | 0.050 ± 0.024 | 0.046 ± 0.001 |
| | 0.032 ± 0.014 | 0.046 ± 0.001 |
| High uranium level with "natural" isotopic ratio | 0.045 ± 0.003 | 0.046 ± 0.001 |
| DU contaminated | 0.012 ± 0.001 | 0.012 ± 0.001 |
| | 0.012 ± 0.001 | 0.013 ± 0.001 |
| | 0.014 ± 0.002 | 0.014 ± 0.001 |
| | 0.015 ± 0.003 | 0.016 ± 0.001 |
| | 0.013 ± 0.002 | 0.016 ± 0.001 |
| | 0.013 ± 0.001 | 0.013 ± 0.001 |

The soil characterized in TABLE I as low uranium level had approximately 2 ppm, and the ^{238}U content were inferred from the ^{226}Ra plus daughters and from the 63.3 keV gamma ray line. By doing so, the gamma counting statistical errors were large, as reflected in the uranium isotopic ratio, which is a function of the uranium content of the soil. This can be seen, by observing in TABLE I, the results obtained by gamma spectrometry for low and high uranium levels with "natural" isotopic ratio. In addition, in the samples measured ^{238}U was not in equilibrium with its long-lived daughters ^{234}Th and ^{226}Ra . As a consequence, the only ^{226}Ra based U ratio measured, considered to be "natural," was that of the soil with high uranium content, i.e. 0.045. This isotopic ratio is consistent with the results obtained by means of mass spectrometry for soils with low and high uranium levels with "natural" $^{235}\text{U}/^{238}\text{U}$ isotopic ratio. The agreement obtained for the results of the measurements of DU contaminated soil, made either by gamma- or mass spectrometry, indicate that the lower the ^{235}U content in the soil sample the better the agreement between the results obtained with the two techniques will be. One possible explanation for this effect may lie in the fact that the quantitative determination of ^{235}U was based solely on the 185.72 keV gamma ray resulting from the alpha decay of ^{235}U to ^{231}Th , which has an emission probability of 57.2%. The interference of the 186.2 keV gamma ray resulting from the ^{226}Ra alpha decay to ^{222}Rn , both pertaining to the ^{238}U series, in the determination of ^{235}U is a known fact in gamma spectrometry of soils with naturally occurring radionuclides. Thus, a correction for the ^{226}Ra content of the sample is usually made by taking into consideration the more intense gamma rays from the ^{226}Ra daughters, such as: 295.2 keV (18.2%), and 351.9 keV (35.1%) from ^{214}Pb ; and 609.3 keV (44.6%), 1120.3 keV (14.7%), and 1764.5 keV (15.1%) from ^{214}Bi . Such type of correction works properly when radon gas (^{222}Rn) does not escape from the soil sample while being measured. This is the main shortcoming of such corrective procedure.

Gamma spectrometric measurements have been improving since the early seventies. However, the

methodology to measure the $^{235}\text{U}/^{238}\text{U}$ isotopic ratio by means of gamma spectrometry were maintained very similar to that described above, which was introduced in ref. [10], but it is still in use today [3-8], irrespective the improvements in gamma detection and analysis systems.

3. THE DETECTION SYSTEM

The detection system is made of a HpGe, able to detect photopeaks at photon energies as low as 30 keV. The detector is inside a shadow shield with 2.0 m length x 1.5 m height x 1.0 m width made of 18 metric tonne of steel fabricated before the World War II, thus free from radioactive contamination from the nuclear weapons tests fallout. The ceiling and the floor have each a thickness of 0.20 m, while the walls are 0.10 thick. The interior of the shielding is covered with 1.0 mm thick copper sheets to absorb x-rays emitted from the shield. Figure 1 is a schematic representation of the internal lateral view of the detection system. The stainless steel chamber is used only when the radon can affect considerably the counting results. The shadow shield is located in a subsurface laboratory room. The air exchange rate between the laboratory room and the exterior is kept to a minimum through a series of barriers. Access to the room housing the shielding can only be made through an entrance tunnel and a glass antechamber. The purpose of the barriers is to decrease the radon concentration in the measuring room. As a consequence, the K_{α} and K_{β} Bi x-rays (74.8, 77.1; 87.2, 89.8 keV), respectively, and the photopeaks of ^{214}Pb (242, 295, and 352 keV), and of ^{214}Bi (609, 768, 934, 1378, and 1765 keV) are absent from the background spectra, making it almost flat in the energies lower than 100 keV.

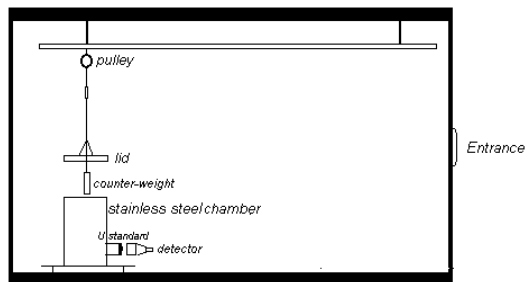


Figure 1. Internal lateral view of the detection system described in greater detail elsewhere [9].

4. METHODOLOGY

The energy calibration of the detection system was made by using the multi-gamma ^{166m}Ho procedure described elsewhere [11]. The isotopic ratio $^{235}\text{U}/^{238}\text{U}$ can be successfully determined by means of estimating the ratio of the areas under the photopeaks of 185.72 keV, with an emission probability of 57.2 %, over 63.29 keV (^{238}U), which has a probability of 3.8 %. However, the ^{226}Ra photopeak at 186.21 keV with 3.5 % interferes with that of ^{235}U at a near energy (i.e., 185.72 keV). This makes mandatory to subtract the ^{226}Ra contribution from the area

under the 185.72 keV plus 186.21 keV photopeaks, whenever one is measuring environmental or biological samples, because in most cases these two photopeaks are not resolved in the spectra.

This paper uses the area under the 143.76 keV ^{235}U photopeak (10.96 %) instead of the 185.73 keV (57.2 %), avoiding this way the need for subtracting the ^{226}Ra contribution from the 186.2 keV (3.5 %).

One additional advantage of this procedure in association with the low gamma counting system described earlier is that photopeaks of fission products may appear clearly in the gamma spectra of environmental or biological samples measured, indicating whether DU contamination may come from an enrichment, or a reprocessing plant. Depending on the $^{235}\text{U}/^{238}\text{U}$ isotopic ratio in the contaminated material, one can also determine if it is NU, or in some rare cases LEW.

Figure 2 represents a graphic of the $^{235}\text{U}/^{238}\text{U}$ isotopic ratio of the areas under the photopeaks at 143.76 keV and 63.29 keV, respectively, as a function of the $^{235}\text{U}/^{238}\text{U}$ standards for 0.31 % (DU), 0.72 % (NU), 1.95 % (LEW), and 2.95 % (LEW). The statistical errors, based on the Poisson distribution of the counts, vary from 14% for the lower standard percent ratio (0.31%) down to 9% for the highest one used (2.95%). The linear fit of the data has a correlation coefficient of 0.9948.

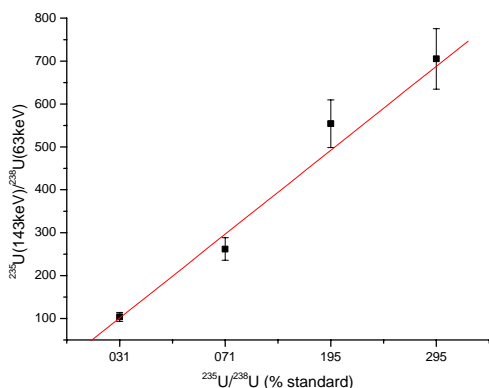


Figure 2. Graphic representing the linear fit calibration of the $^{235}\text{U}/^{238}\text{U}$ ratio based on the counting areas under the photopeaks at 143.76 keV and 63.29 keV, respectively, as a function of the $^{235}\text{U}/^{238}\text{U}$ standards for DU, NU, and LEW.

The calibration curve presented in Figure 2 was rather preliminary, with only 300 seconds counting time for each of the four standards. Longer measurements will be made to improve the counting statistics, taking into account that the counting system is rather stable up to times of the order of 5×10^5 seconds [9].

Soil samples contaminated with DU were not available to us for measuring at the time of this writing. The calibration of the detection system using the procedures described here eliminates the need for correction for the non-equilibrium between ^{238}U and ^{226}Ra in environmental and biological samples, as it is usually the case. An intercalibration exercise to compare the methodology described in this work, that means, using the 143.76 keV

^{235}U photopeak (10.96 %) instead of the 185.73 keV to measure the $^{235}\text{U}/^{238}\text{U}$ ratio is rather desirable.

5. CONCLUSIONS

1. The feasibility of calibrating a low gamma counting detection system with a shadow shield for measuring DU, NU, and LEW has been demonstrated.
2. The main advantage of using the ^{235}U 143.76 keV photopeak, instead of the more commonly used 185.72 keV, for measuring the $^{235}\text{U}/^{238}\text{U}$ ratio resides in avoiding the need to subtract the ^{226}Ra contribution in environmental samples.
3. This calibration procedure is particularly helpful when one needs to measure the $^{235}\text{U}/^{238}\text{U}$ ratio in environmental and biological samples

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