An alternative method to determine ^{235}U in environmental samples

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Abstract. In this work, three different procedures (direct gamma-ray spectroscopy, indirect gamma-ray spectroscopy and gamma-gamma coincident spectroscopy) are employed in the determination of the amount of ^{235}U in a well-known enriched-Uranium sample, and all three are discussed as possible alternatives to determine ^{235}U in environmental samples. The results show that the three methods give similar results, although these results are all ~30% below the expected value.

1 Introduction

From the viewpoint of environmental radioactivity monitoring, it is important to measure the Uranium concentration and isotopic ratio at regular time intervals, as this can be used to check for nuclear activities, mainly the operation of nuclear facilities like nuclear power and/or fuel-processing plants.

A quick and low cost way to perform those measurements without any chemical treatment is gamma ray spectroscopy. In these measurements, the direct determination of ^{238}U without any chemical treatment using semiconductor gamma ray spectrometer is very hard because this isotope has no intense gamma rays and a low specific activity (1.2×10^4 Bq $\cdot$ g^{-1}). On the other hand, it has several byproducts which have more intense lines and activities that could be equal to that of their parents if the conditions of secular equilibrium are installed.

A frequent way to measure the ^{238}U using its daughters is by measuring the 63 and 93 keV transitions from ^{234}Th. Another way is by using the 1001 keV line from ^{231}Pa. Nevertheless, ^{235}U in spite of its low isotopic abundance has intense gamma rays; the strongest gamma line is at 185 keV, however there is a transition from the decay of ^{226}Ra, which is in the natural decay series of ^{238}U, with an energy that's very close to this (E = 186 keV) and, despite the low relative intensity of the latter (I = 3.5%), the ^{226}Ra concentration in environmental samples is much greater than that of ^{235}U, thus making the analysis of the 185 keV transition from ^{235}U difficult.

One way to overcome this interference is by using of a high-resolution detector, fitting the resulting peak as a doublet (185–186 keV) and then using the 143 keV transition from ^{235}U to assert the quality of the fit. The measurement of this transition is tricky, though, due to its much lower intensity ($I_\gamma = 11\%$); also, in this lower-energy region many secondary effects of higher energy transitions – as backscattering or Compton scattering – may interfere in the measurement. Another possibility is to check the ^{235}U activity indirectly by measuring the 1001 keV transition from the decay of 234Pa, thus avoiding the problems related to the low-energy region of the spectrum.

The present study aims to determine ^{235}U in a well-known sample using coincident gamma spectroscopy, so that this doublet can be properly separated analyzing the coincidences of the 185 keV transition with the 202 keV transition, thus eliminating the undesired contributions from other nuclides. These results will then be compared with the results from a conventional single-gamma spectroscopy analysis both in the 143 and 185 keV transitions from the decay of ^{235}U and in the 1001 keV transition from the decay of ^{234}Pa, allowing for a discussion on the performance and applicability of these methods.

2 Experimental procedure

For this work, 7.258 g of 20%-enriched uranium oxide were disposed into a cylindrical plastic container (internal dimensions: 40.6 mm diameter, 3.5 mm thickness). Two HPGe detectors with active volumes 76 cm^3 and 89 cm^3 were placed face to face, at a distance of 10 cm, and the sample is placed, aligned with the detectors' axes, in the middle of this distance. For the single-gamma spectroscopy measurements, the 89 cm^3 detector was used and, for the coincidence measurements, the 76 cm^3 detector was used as a gate while the acquisition was performed by the 89 cm^3 detector (see fig. 1). Both detectors were independently calibrated in energy and efficiency using ^{60}Co, ^{152}Eu, ^{133}Ba and ^{137}Cs standard sources; the coincidence system was calibrated using the ^{60}Co source (gated on the 1332 keV line), the ^{152}Eu source (gated on the 244 keV line) and the ^{133}Ba source (gated on the 357 keV line).

The efficiency function used was:

$$Eff(E) = \left(a_1 \cdot e^{(a_2 \cdot E)} + a_3 \cdot e^{(a_4 \cdot E)} \right) \cdot e^{\left(\frac{\mu}{\rho} \cdot x \right)} \tag{1}$$

where a_1, a_2, a_3 and a_4 are parameters to be fitted, μ/ρ is the germanium mass attenuation coefficient [1], and x is a parameter associated with the detector dead layer, also to be fitted. An attempt was made to estimate experimentally the attenuation due to the self-absorption in the sample by placing the calibration sources behind the sample, but the fit of these data was very tricky because there's an absorption
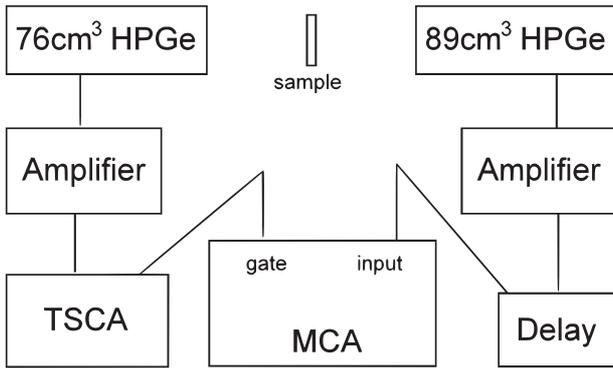
Fig. 1. Schematic drawing of the experimental setup.

edge around 111 keV due to k_β of uranium, which cause the efficiency function not to have a smooth energy dependence in the region of interest, thus making a fit of this function practically impossible.

The activity of gamma-emitting nuclides in the ^{238}U and ^{235}U chain were calculated in the single spectroscopy data by

$$Activity(cps) = \frac{Counts}{T \cdot Eff(E) \cdot I_\gamma} \qquad (2)$$

where *Counts* is the total area of the photopeak of gamma energy E, T is the time of acquisition, I_γ is the transition intensity and *Eff(E)* is the absolute detection efficiency calculated from the energy–efficiency relation. To transform the activity of ^{238}U to ^{235}U, the relation $^{238}U/^{235}U = 4$ was used.

In the gamma-gamma coincidence measurements, the efficiency was calculated by

$$Activity(E) = \frac{Counts}{T \cdot I_\gamma \cdot br \cdot \varepsilon_1 \cdot \varepsilon_2 \cdot W(\theta)} \qquad (3)$$

where T is the time of acquisition, I_γ is the emission probability of the first photon of the cascade br is the normalized branching ratio of the second photon of the cascade. $W(\theta)$ is the angular correlation function for the angle between the symmetry axes of the detectors.

The accidental contributions were estimated by the same way but changing the branching ratio by the emission probability of the second photon, and the angular correlation by a constant related to accidental events.

3 Results and discussion

The single gamma spectroscopy was recorded over 19 hours of live counting and the background was measured in the same conditions.

The expected activity in the sample due to ^{235}U can be calculated as

$$A = \lambda N \qquad (4)$$

Table 1. ^{235}U activity estimated using the three methods proposed in this work.

Method	Transition	Activity (kBq)
Direct determination	185 keV	73.769 (25)
	143 keV	69.038 (35)
Indirect determination	1001 keV	69.97 (28)
Coincidence (gate in 185 keV)	202 keV	65.30 (23)

where N is the number of ^{235}U atoms in the sample, given by

$$N = \frac{mN_a}{M} \qquad (5)$$

where m is the sample mass, N_a is Avogadro's constant and M the atomic mass of ^{235}U. The resulting expected activity is 106 kBq.

Also, although the results could not be experimentally corrected for the self-absorption in the sample, this attenuation can be estimated as:

$$\frac{\int_0^R e^{\mu \cdot x} \cdot dx}{\int_0^R dx}. \qquad (6)$$

As the density ρ of the sample was calculated as 4 g/cm^2 and the depth is R = 0.35 cm, if we use an average attenuation coefficient μ/ρ, for the energy range from 150–200 keV, of ~2 cm^2/g [1], we come to an approximate attenuation, in this energy range, of 34%.

Applying the attenuation coefficient estimated above to the expected activity of ^{235}U, one finds that the expected "absorption-corrected activity" for the 150–200 keV region would be 70 kBq, which is roughly compatible with the results obtained for the direct single spectroscopy measurements; the results obtained for the coincidence method are somewhat lower, but this could also be a consequence of the self-attenuation, as this can be expected to be harder to estimate for this measurement. For the indirect measurement, though, the attenuation coefficient is much lower (~1%), so the results obtained by this method fall quite below the expected value.

4 Conclusion

The three methods used to determine ^{235}U activity present some difficulties. The direct method, besides being affected by the low quantity of ^{235}U in environmental samples, is also problematic due to the fact that the most intense transitions (185 and 143 keV) are in the lower-energy region where many secondary effects of higher energy transitions – as backscattering or Compton scattering – may interfere in the measurement, and where the self-absorption in the Uranium of the sample can be of significance. The indirect method also presents some difficulty, because the time to install the secular equilibrium is at least seven half-lives of the element with smaller decay constant after the father. The coincidence method, although even more affected by the small presence of ^{235}U in environmental sources, is free of most low energy

effects. In this work the three methods were compared and the results were smaller than the expected values. In the coincidence and in the direct methods, the principal limitation was the difficulty to consider the self-attenuation effects. In the indirect method, due to the manipulation of the U sample, it is possible that the conditions of secular equilibrium in the ^{238}U family related to our goal (up to ^{234}Pa) were not established. Part of aim of this work was to propose the measurement of coincidence with the simplest experimental setup possible, however these results show that a new evaluation with an improved arrangement shall be made; also, measurements with a thinner source may help in reducing the self-absorption effects, although at the cost of reducing also the sample's activity.

References

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