ACTIVITY DETERMINATION OF ^{238}U AND ^{230}Th BY GAMMA SPECTROMETRY TO METROLOGICAL DEVELOPMENT

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Abstract: This work describes the difficulty for metrological determining of the activity of ^{238}U and ^{230}Th using gamma spectrometry due to the low gamma-ray emission probabilities of 92 and 67 keV, and, mainly, the associated high uncertainties, 13 and 11%, respectively. ^{230}Th is a ^{238}U daughter and it is product from uranium mills. The natural radionuclide ^{230}Th decay to ^{226}Ra, and the transition has to be measured because there are not secular equilibrium between these radionuclides and daughters.

Key words: ^{238}U, ^{230}Th, germanium detectors, efficiency curves

1. INTRODUCTION

The control of fissionable material as ^{238}U, is important because ^{239}Pu is produced in reactors as a result of origin neutron capture by ^{238}U to produce ^{239}U, with beta decays first to ^{239}Np then to ^{239}Pu. ^{239}Pu can be used in thermal reactors as a mixed fuel to reactors and others applications. [1].

^{230}Th is a member of ^{238}U decay chain (alpha decay). It is found in natural thorium when uranium is present in thorium ores. It has a long half-life (80,000 years) and its activity remains practically constant. Thorium can be also used as a fuel reactor [2].

The gamma-ray measurement methods are widely applicable to bulk fissionable materials [3] . By the detection efficiencies for a particular geometry the activity of a source can be determined by gamma spectrometry. The sources can be liquid or solid form. Also, the high purity germanium detector (HPGe) has an excellent resolution, a good separation of gamma-ray peaks, and it is used in the radionuclide identification.

The determination of the activity of a radioactive source needs the detector efficiency response knowledge in function of the energy. The efficiency calibration establishes the relationship between photopeak area and the nuclide activity as:

$$\varepsilon_f(E) = \frac{S(E)}{[A.I(E).t]} \qquad (1)$$

Where:
$\varepsilon_f(E)$ is the total absorption efficiency at E energy;
A is the activity (Bq);
$S(E)$ is the peak area (counts);
$I(E)$ is the absolute probability emission of the energy considered for the specific nuclide;
t is the counting time.

The total absorption efficiency determination can be done from geometric considerations and interaction probabilities or using a semi-empirical relation [4]. To determination of the detector response are used standards with a large energy range, or a multi-gamma and x-ray emitter standards like ^{166m}Ho (48 to 1000 keV) [4,5], ^{152}Eu (100 to 1400 keV) [5,6] , ^{133}Ba (30 to 400 keV) [5] and ^{57}Co (14 to 140 keV) [5] according to the region of interest. These radionuclides can be used to build an efficiency curve.

The efficiency curve depends on radiation energy [7], sample geometry, photon attenuation (sample absorption and material absorption between sample-detector), dead time and sample-detector position. This curve is obtained from the acquisition of reference spectra considering the photopeak areas corresponding the standard activities.

2. PURPOSE

From a high-resolution germanium detectors and their well-fitted efficiency curves in a defined geometry and a determined range of energy is possible to do a metrological sample calibration in activity, uncertainties included. When the laboratory of calibration or essay does not own a standard of same nature of the sample, the efficiency curve method should be used instead of the commonly comparative method [4,8].

In this work, it was used the experimental arrangements for measuring precisely the activity and associated uncertainties of gamma-emitter point and bulk sources using the efficiency curve method to the samples of ^{238}U and ^{230}Th. The choice of photopeak energy is a difficult task to proceed the analysis.

3. METHODOLOGY

For the activity analysis purpose were determined efficiency curves in a set source-detector distance with multi-gamma acrylic point source standards of ^{152}Eu, ^{133}Ba and ^{166m}Ho and liquid source standards (ampoules) of ^{57}Co and ^{133}Ba with traceability to the international reference system [4]. In this methodology it was adopted a 5 cm distance between detector-source in order to minimize pile-up losses and source geometry effects [6]. A general schematic view of gamma-ray spectrometric systems with HPGe detectors is shown in Figure 1.

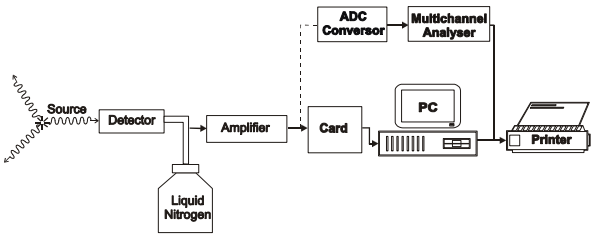

Fig. 1. Gamma-ray spectrometric system with germanium detector.

The first detector consists of an Ortec HPGe coaxial detector (GEM50P4), with a volume of 260 cm3, a resolution (FWHM) of 1.90 keV at 1332.5 keV and a 66:1 Peak-Compton ratio. The other detector consists of a Canberra HPGe planar detector (Gl2020R), with a volume of 40 cm3 and a resolution (FWHM) of 6.4 keV at 122 keV.

The acrylic source to be analysed was prepared by dropping the original solution on acrylic disc. After drying, this sample was covered with other disc and sealed. This acrylic source has the following dimensions: 25.4 mm of external diameter, 5.0 mm of height, 1.0 mm of thick for deposition area and 1.0 mm of covering. The liquid source is a glass ampoule, it has the following dimensions: 5mL of capacity, 90 mm of height,14 mm of external diameter, 0.5 mm of thick, and the sample solution has 2.6-2.7 g that corresponding of 20 mm of solution height.

To activity determination of ^{238}U acrylic source the gamma energies 63.29 keV (4.5 %) and 92.59 keV (5.2%) [9-11] , from its first progeny, ^{234}Th, were studied, but it was only considered the 92.59 keV photopeak because of the lower associated uncertainties of emission probabilities (about 13%) when compared with other energy (about 20%). It was used the coaxial detector (Figure 2).

To determining ^{230}Th ampoule source activity was used the Ge planar detector and its correspondence efficiency curve can be seen in Figure 3. The energy of photopeak considered to do this analysis was 67.73 keV with 11,4% [9,10] of uncertainty .

4. RESULTS AND DISCUSSION

Efficiency curves (Figures 2 and 3) obtained as well as the results related to activity determination with associated uncertainties for the following radioactive samples: ^{238}U acrylic source and ^{230}Th ampoule source (Table 1). The main uncertainty contributions come from the efficiency interpolation (~1.3%), the peak area determination (~0.2%) and uncertainties of emission probability among others. The choice of 5cm source-HPGe detector distance was due to sample sources low activities.

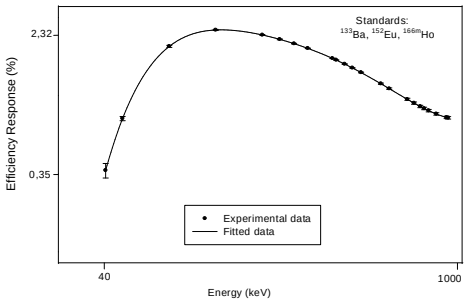

Fig. 2. Efficiency curve obtained with ^{133}Ba, ^{152}Eu and ^{166m}Ho acrylic standard sources. The distance of source-HPGe coaxial detector (GEM50P4) is 5 cm.

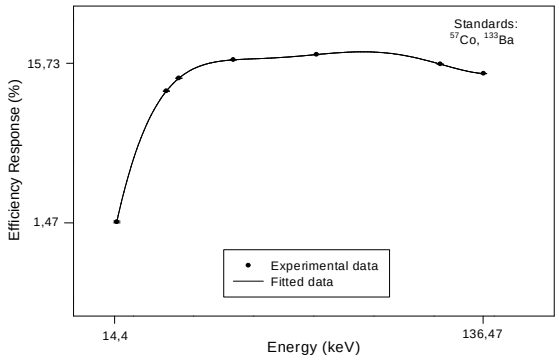

Fig. 3. Efficiency curve obtained with ^{57}Co and ^{133}Ba liquid standard sources. The distance of source-HPGe planar detector (Gl2020R) is 5 cm.

Table 1. Activities and uncertainty (U) values for radionuclides.

Nuclide	Certificated Activity (Bq)	Measured Activity (Bq) ± U (%)
^{238}Uacrylic (a)	_ (c)	37.2 ± 13.7
^{230}Th ampoule (b)	124.4 ± 0.9 (d)	120.3 ± 11.7

(a) coaxial germanium detector; (b) planar germanium detector (c) unknown certificated value; (d) liquid cintilation

The activity of ^{238}U sample was determined from the peak area counting related to 92.59 KeV energy and from the efficiency curve (Figure 2). No significant impurities have been observed. Using the efficiency curve of HPGe coaxial detector the result of ^{238}U activity was 37.2 Bq with 13.7 % of uncertainty. The certificated value is unknown to this source, then, it was not possible to do the comparison.

The activity of ^{230}Th was determined from the efficiency curve (Figure 3) and the result (120.3 Bq ± 11.7%) showed 3.2% of deviation in comparison with the certificated value. In this work no significant impurities have been observed.

Due to the low activity of radionuclides present in environmental samples, it is difficult to get a good count statistics.

5. CONCLUSION

This work allows obtaining metrological results to radioactive point and bulk sources of low activity by means of Ge detectors using the efficiency curve method. The uncertainty of 1σ was considered. The results uncertainties are firstly from low gamma-ray emission probabilities. The choice of photopeak energy was a difficult task to proceed the analysis.

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