Impurity determination in ^{153}Sm by means of a High Purity Germanium spectrometer

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Resumo: Neste trabalho, a impureza radionuclídica de longa duração ^{152}Eu, presente na solução de ^{153}Sm, foi medida por meio de um espectrômetro de HPGe, calibrado na faixa de energia entre 81 e 1408 keV por meio de soluções padronizadas em ampolas seladas de ^{60}Co, ^{133}Ba, ^{137}Cs e ^{152}Eu. A razão entre as atividades do ^{152}Eu e do ^{153}Sm resultou 5,2x10^{-6}. Este valor é menor que o limite estabelecido pelo controle de qualidade.

Palavras-chave: Espectrometria Gama, Samário-153, Európio-152.

Abstract: In the present work, the long-lived radionuclide impurity ^{152}Eu, present in ^{153}Sm solution was measured by mean of a HPGe spectrometer, which was calibrated in the energy range between 81 and 1408 keV by means of standard sealed ampoules of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu. The activity ratio between ^{152}Eu and ^{153}Sm resulted 5.2x10^{-6}. This value is lower than the limit established by the quality control.

Keywords: Gamma spectrometer, Samarium-153, Europium-152.

1. INTRODUÇÃO

This work aims to determine long-lived radionuclidic impurities present in ^{153}Sm solution by means of a High Pure Germanium spectrometer, to meet the requirements for radiopharmaceutical quality control.

This radionuclide can be produced in nuclear reactors by means of ^{152}Sm(n,γ)^{153}Sm reaction and it is used in nuclear medicine as Samarium-153 ethylenediaminetetramethylenephosphonic acid ([^{153}Sm]EDTMP) to palliate pain resulting from osteoblastic metastatic bone cancer and, also the intractable skeletal pain due to disseminated body metastases [Singh, 1989]. It decays with 1.92855(5) d half-life, emitting three main beta particles with endpoint energies of 634.7 keV 704.7 keV and 807.6 keV, with probabilities of 34.4%, 49.2% and 19.5%, respectively, and a main gamma ray of 103.18 keV [Bé, 2006].

For this measurement, a high purity germanium spectrometer was calibrated in the energy range between 81 keV and 1408 keV by measuring standard sealed ampoules of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu, standardized by the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear - LMN) of the Nuclear and Energy Research Institute - IPEN in São Paulo.

The natural occurring samarium has seven stable isotopes composed by ^{144}Sm, ^{147}Sm,

^{148}Sm, ^{149}Sm, ^{150}Sm, ^{152}Sm and ^{154}Sm with isotopic percentage of 3.08(1)%, 15.02(5)%, 11.22(3)%, 13.83(9)%, 7.35(2)%, 26.74(3)% and 22.73(5)%, respectively [Changa, 2002]. Due to this composition, the irradiation in the reactor, besides the radionuclide of interest, may produce a series of other radioisotopes, so that the solution of interest shows many radionuclides. To avoid the presence of these contaminants, enriched targets are used.

In this work, the main impurity detected was ^{152}Eu, which is formed by ^{151}Eu (n,γ)^{152}Eu reaction in elemental ^{151}Eu impurity. The ^{152}Eu decays by 72.1%, by electron-capture process, and by beta minus emissions (27.9%), emitting multiple high energy gamma rays in the range of 121 keV to 1408 keV [Vanin, 2004].

2. EXPERIMENTAL METHOD

2.1. Sample preparation

The ^{153}Sm supplied for this work by the IPEN Radiopharmaceutical Center was produced by thermal neutron irradiation of isotopically enriched 98.44(±0.1) % $^{152}Sm_2O_3$ targets. In table 1 the isotopic distribution of the other samarium isotopes provided by the supplier certificate is presented and in table 2 the elemental composition of the impurities in parts per million (ppm) are presented [ISOFLEX USA, 2013]. The sample used for this measurement was prepared in flame sealed ampoule, with 1 mL from a $^{153}SmCl_3$ solution diluted in 0.1 N HCl.

Table 1. Isotopic distribution provided by the supplier certificate.

Isotopic Distribution (%)					
^{144}Sm (0.01)	^{147}Sm (0.06)	^{148}Sm (0.08)	^{149}Sm (0.14)	^{150}Sm (0.24)	^{154}Sm (1.03)

Table 2. Elemental composition of the impurities in parts per million (ppm) from the supplier certificate.

Elemental Composition (ppm)		
K (<50)	Na (<20)	Ca (<50)
Mg (<3)	Fe (<50)	Al (3)
Si (<50)	Ni (<1)	Cu (2)
Pb (3)	Sn (<1)	Zn (2)
Pt (71)	Yb (1)	Gd (9)
Eu (<60)	Nd (<1)	Pr (<1)

2.2. Gamma-ray spectrometry measurements

The gamma-ray measurements were performed in a REGe coaxial detector with 500 μm thick Be window, yielding a resolution of 1.79 keV FWHM at 1332.5 keV. This detector was previously calibrated in the energy range between 81 keV and 1408 keV, by measuring sealed flamed 5 mL Schott ampoules with 13.3 mm in diameter containing ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu solutions standardized by the LMN. These ampoules are traceable to standard sources supplied by the International Atomic Energy Agency (IAEA). The variation in height was estimated and the geometry and attenuation factors were calculated accordingly. The variations in these factors were included in the overall uncertainty.

The source-detector distance was around 17.9 cm to minimize the sum coincidence effect. The standard ampoules and the ^{153}Sm ampoule were positioned in a Plexiglass holder. Dead time and pile-up corrections were applied by measuring a reference pulser peak near the upper edge of gamma spectrum, together with the radioactive sources. The total absorption peak area was determined by simple spectrum integration with linear background subtraction from both sides of the peak, applying ALPINO code [Dias, 2001].

The ^{153}Sm activity and ^{152}Eu imputiry activity were determined by using the following equation:

$$A = \frac{S(E_\gamma)}{\varepsilon(E_\gamma)\, p(E_\gamma)\, T\, f} \quad (1)$$

Where:

ε (E_γ) gamma ray peak efficiency of energy E

S(E_γ) Area under the total absorption peak of gamma ray of energy E determined by the Alpine code.

A Source activity.

T Measurement time.

f Correction factors (dead time, attenuation, geometry).

p(E_γ) emission probability per decay of gamma ray of energy E.

3. RESULTS

The efficiency curve for the coaxial REGe detector was fitted by the least squares method using LOGFIT code (Dias and Moreira, 2005), which considers covariance methodology in order to determine all uncertainties. The reduced $\chi 2$ value was close to 1, indicating a satisfactory fit.

The calibration curve was fitted by a forth order polynomial function in log-log scale. The full energy peak efficiency curve as a function of the gamma-ray energy is presented in figure 1. The percent residuals between experimental and fitted efficiencies are shown in figure 2. The fitting parameters are presented in table 3.

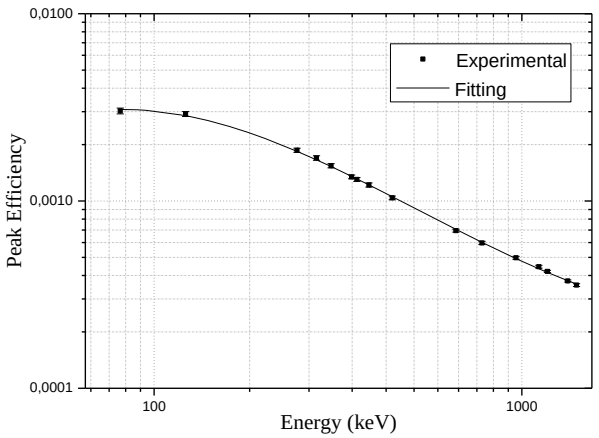

Figure 1. Full energy peak efficiency of the REGe coaxial detector. The black marks correspond to experimental points and the continuous line to fitting.

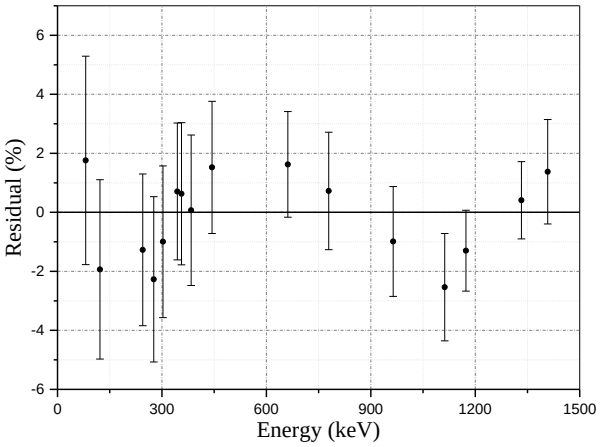

Figure 2. Percent residual between experimental and fitted efficiencies.

Table 3. Fitting parameters with the covariance matrix.

Parameters	Value	Uncertainty	Covariance Matrix			
a_0	-24.82	2.1	4.44			
a_1	10.57	1.1	-2.29	1.19		
a_2	-1.85	0.2	0.39	-0.20	0.03	
a_3	0.09	0.01	-0.02	0.01	-0.02	0.0001

The ^{153}Sm activity and ^{152}Eu imputiry activity were obtained, by the analysis of the spectrum measured 30 days after the date of calibration; the counting time was 200,000 s. In figure 3, the ^{153}Sm ampoule spectrum is presented: in this spectrum, the main gamma rays from ^{152}Eu are indicated. As may be seen in this spectrum, the presence of ^{154}Eu at 1274 keV, which is one most intense gamma-ray emitted (34.9%), is quite low. Nevertheless, the impurity level is lower than the quality control requirement, which shows that the ^{153}Sm solution is in accordance with the Good Manufacturing Practices and Control, required by the Brazilian National Surveillance Agency, from the Ministry of Health (ANVISA).

Further measurements have been carried out to verify the presence of ^{154}Eu and other radionuclides impurities. A measurement using a lead shield to minimize the ^{153}Sm activity contribution is in progress in order to enable the measurements at the date of calibration.

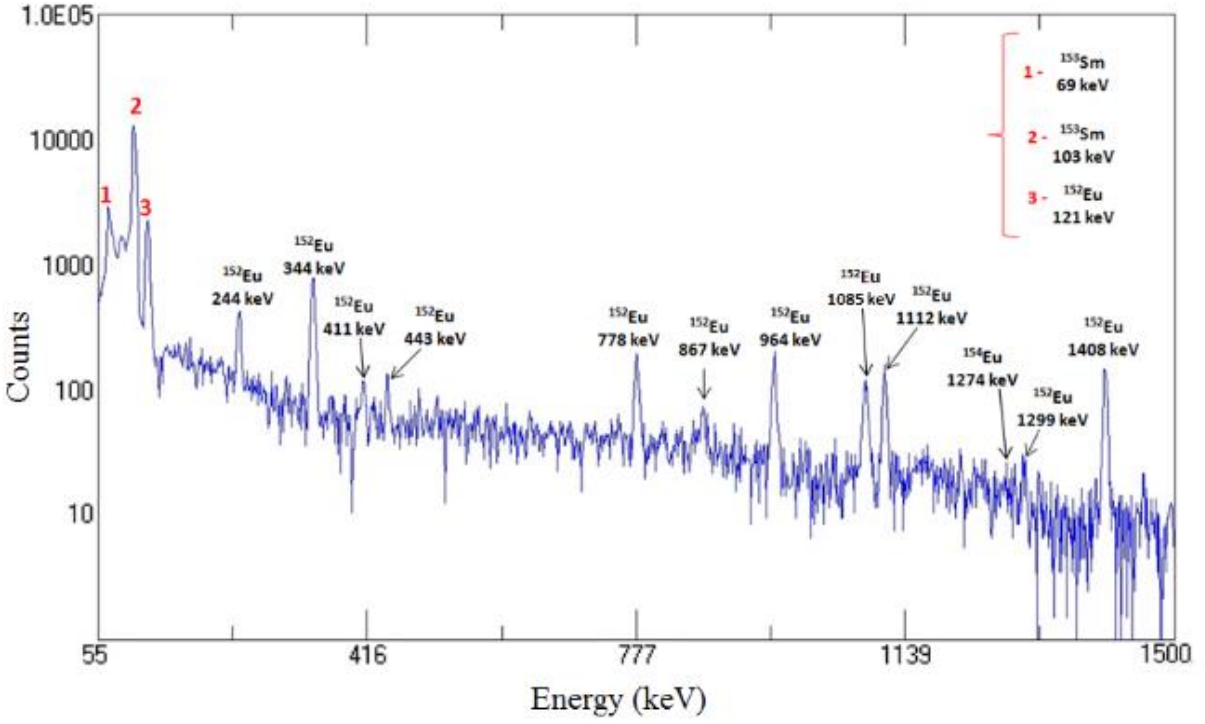

Figure 3. ^{153}Sm ampoule spectrum.

The weighed mean activity of the ^{152}Eu impurity and the activity of ^{153}Sm obtained from the ampoule measurement at the calibration time were (622 ± 17) Bq and (119 ± 5) MBq, respectively, giving a ratio of (5.2 ± 0.3) x10^{-6}, which is lower than the limit required by regulations.

4. CONCLUSION

The ^{152}Eu was identified as the main impurity present in the ^{153}Sm solution analysed.

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