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Determination of ⁵¹Cr and ²⁴¹Am X-ray and gamma-ray emission probabilities per decay

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ARTICLE INFO	ABSTRACT		
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Emission probability per deca Americium 241 Chromium 51 Semiconductor detectors In this paper results of X-ray and gamma-ray emission probabilities per decay of ⁵¹Cr and ²⁴¹Am are presented. The measurements were carried out by means of HPGe planar and REGe spectrometers. The activity of ⁵¹Cr and ²⁴¹Am samples was determined in a $4\pi\beta - \gamma$ coincidence counting system. The HPGe spectrometers were calibrated in a well defined geometry by means of ⁵⁴Mn, ⁵⁵Fe, ⁵⁷Co, ¹³³Ba, ¹⁵²Eu, ^{166m}Ho and ²⁴¹Am sources, previously standardized in a $4\pi\beta - \gamma$ coincidence system. The MCNP Monte Carlo code was used for simulation of the REGe spectrometer calibration curve, for the selected geometry, and compared with the experimental curve. The experimental results were compared with data from literature.

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1. Introduction

Radionuclides which decay by X-ray and gamma-ray emission are used in several applications such as agriculture, medicine, environment, especially with semiconductor detectors. However, in order to use these gamma-ray emitters it is necessary to have their parameters well known, such as: half-life, energy and mainly X-ray and gamma-ray emission probabilities per decay. These latter parameters have presented discrepancies in literature values, especially in the low energy region, due to lack of standards and complex spectra. In the low energy region, there are many multiplets that have to be solved by deconvolution methods and secondary effects, which have to be considered. In this paper, two radionuclides which present discrepancies in the emission probabilities values, namely ⁵¹Cr and ²⁴¹Am, were selected to be measured.

Radionuclide ⁵¹Cr is widely used in nuclear medicine. It decays 9.89% by electron capture to the excited states of ⁵¹V, followed by a 320.08 keV gamma-ray emission, and 90.11% by electron capture to the ground state of ⁵¹V (Bé et al., 2004).

²⁴¹Am decays with a half-life of 432.2 y (Bé et al., 2004), emitting alpha particles populating the excited levels of ²³⁷Np followed by several low energy gamma-rays. The radionuclide ²⁴¹Am was also selected because the emission probabilities of the 26.34 keV gamma-ray and X-rays present large discrepancies in the literature.

The activity of 51 Cr and 241 Am samples, used in this paper, was determined by means of a $4\pi\beta-\gamma$ coincidence counting system.

The emission probability was measured in two HPGe spectrometers, a coaxial detector for the gamma-ray and a planar detector for X-ray and low energy gamma-ray measurements. The spectra analysis was performed by the codes COLEGRAM (Ruellan et al., 1995, 1996) and ALPINO (Dias, 2001).

2. Experimental method

2.1. HPGe spectrometers

The REGe coaxial detector presents 1.79 keV FWHM at 1332.5 keV, and the HPGe planar 160 eV FWHM at 5.9 keV. The detector windows were made of Be, 500 and 50 µm thick, and the source-detector distance was 17.9 cm and 10.4 cm, respectively. These spectrometers were calibrated using standard sources, previously calibrated in a $4\pi\beta - \gamma$ primary system in the Nuclear Metrology Laboratory (LMN) at IPEN.

The 241 Am solution used in this work was sent by the Bureau International des Poids et Mesures (BIPM) to the LNMRI, in Rio de Janeiro, which forwarded a fraction to our laboratory in São Paulo. The activity was measured in a $4\pi\alpha - \gamma$ coincidence system (Koskinas et al., 2006). The 51 Cr was supplied by the Radiopharmaceutical Center of IPEN, the activity was determined in a $4\pi(X, A) - \gamma$ coincidence system.

The samples for spectrometer calibration and for the gamma intensity measurements were prepared by dropping known aliquots of the solution on a $20 \,\mu g \, cm^{-2}$ thick Collodion film. This film had been previously coated with a $10 \,\mu g \, cm^{-2}$ gold layer in order to render the film conductive. A seeding agent (CYASTAT SM) was used for improving the deposit uniformity and the sources were dried in a dessicator. The mass determination was

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Fig. 1. (a) Full energy peak efficiency of the HPGe planar detector and (b) percent residues between experimental and fitted efficiencies.

performed using the pycnometer technique (Campion, 1975). From ⁵¹Cr and ²⁴¹Am solutions four and six samples were prepared, respectively.

2.2. Spectrum analysis

The gamma-rays and X-rays spectra obtained for the efficiency curve and for emission probability determination were analyzed by COLEGRAM code. For gamma-ray peaks a gaussian distribution was fitted and for X-rays peaks, a Voight profile fixing the Lorentzian width was utilized, in accordance to the natural KXrays lines width. In both analyses, an exponential background was considered, after the experimental background subtraction had been applied. The curve fitting to experimental data was performed by the least square method.

The spectra obtained using the REGe spectrometer was analyzed by Alpino code, which applies the method of simple integration of the total absorption peak by the following function:

$$S(E_{\gamma}) = \sum_{K_2+1}^{K_3-1} C_i \cdot S_B F_K$$
(1)

where C_i is the number of counts in channel *i*, F_K is a factor that consider the number of channels under the absorption peak, S_B is





Fig. 2. (a) Full energy peak efficiency of the REGe coaxial detector and (b) percent residues between experimental and fitted efficiencies.

the background counting rate given by

$$S_B = \sum_{K_1}^{K_2} C_i + \sum_{K_3}^{K_4} C_i$$
(2)

where $K_1 = K_P - 3D$ is the initial channel of background counting, $K_2 = K_P - 2D$ is the initial channel of total absorption peak, $K_3 = K_P + 2D$ is the final channel of total absorption peak, $K_4 = K_P + 3D$ is the final channel of background counting, with K_P the peak centroid and *D* the spectrometer resolution (FWHM).

In both cases, the emission probabilities per decay were calculated by the following equation:

$$p(E_{X,\gamma}) = \frac{S(E_{X,\gamma})}{\varepsilon(E_{X,\gamma})A}$$
(3)

where $S(E_{X,\gamma})$ is the counting rate under total absortion peak of energy $E_{X,\gamma}$, $\varepsilon(E_{X,\gamma})$ is the peak efficiency, *A* is the absolute activity.

2.3. Efficiency curve

The planar efficiency curve was obtained by the analysis of X-ray and gamma-ray in the range between 5 and 80 keV by means of the ⁵⁴Mn, ⁵⁵Fe, ⁵⁷Co, ¹³³Ba, ¹⁵²Eu, ^{166m}Ho and ²⁴¹Am standard sources, prepared according to Section 2.1 and calibrated

Table 1

Emission probabilities per decay of KX-ray and gamma-ray of ⁵¹Cr.

Energy (keV)	This work	Bé et al. (2004)	IAEA (1991)	Fisher and Hersberger (1984)
4.95	0.200(7)	0.2015(30)	0.201(3)	-
5.43	0.0257(9)	0.0269(7)	0.027(1)	-
320.08	0.0987(3)	0.0987(5)	0.0986(5)	0.1030(19)

This values were calculated by averaging individual values from all measured samples. The uncertainties correspond to one standard deviation (u=1).

Table 2

Emission probabilities per decay of 26.34 keV ²⁴¹Am gamma ray.

Energy (keV)	This work	Iwahara et al. (2005)	Bé et al. (2004)	Schötzig and Schrader (1993)
26.34	0.0227(4)	0.0206(3)	0.0240(3)	0.024(1)

The uncertainties are indicated in parentheses and correspond to one standard deviation (u=1).

Table 3 Typical values of the partial uncertainties involved in the emission probabilities per decay determination, in percent (one standard deviation, u=1).

Components	4.95 keV	5.43 keV	320.08 keV	26.34 keV
Mass Statistics Detector efficiency Activity	0.10 3.16 1.64 0.14	0.10 3.17 1.43 0.14	0.10 0.40 0.36 0.14	0.10 0.48 1.50 0.13
Total	3.56	3.48	0.56	1.58

in a $4\pi\beta - \gamma$ coincidence system. This curve was used to analyze the emission probabilities of ⁵¹Cr X-rays and ²⁴¹Am 26 keV gamma-rays. Fig. 1 shows the efficiency curve fitted by the least square method using LOGFIT code (Dias and Moreira, 2005) with the experimental points. The best fit was achieved applying a 2nd degree polynomial in log–log scale, yielding a chi-square value close to one.

To obtain a constant response in this region, corrections for attenuation in Be window and in the air and for escape due the germanium *K*-edge, with theoretical calculation according Debertin and Helmer (1988) were applied.

The REGe coaxial detector efficiency curve was obtained by the analysis of gamma-ray in the range between 59 and 1408 keV using of the 60 Co, 133 Ba, 152 Eu, 166m Ho and 241 Am standard sources calibrated in a $4\pi\beta - \gamma$ coincidence systems. Fig. 2 shows the efficiency curve, used to analyze the emission probability of 320 keV 51 Cr gamma-ray.

A theoretical calculation of efficiency curve for the REGe detector was carried out by means of the Monte Carlo simulation using MCNP-4C code (ORNL, 2001). This curve is presented as a dot line in Fig. 2. As it can be seen, the agreement between experimental and theoretical values is quite good in the medium energy region, in the low energy region there is a difference around 2% for 59 keV of ²⁴¹Am and 6% for 80 keV of ¹³³Ba and ^{166m}Ho. A new calculation with the MCNPX code (McKinney et al., 2006) is underway. This code yields more consistent results when compared to experimental values in the low energy region. A similar efficiency calculation for the planar detector has been initiated for comparison to experimental values.

3. Results and discussion

All ⁵¹Cr and ²⁴¹Am samples were measured in planar or REGe spectrometers, depending on the energy range of interest and the spectra analyzed in order to obtain the emission probabilities.

The averages of all results from KX- and gamma-ray emission probabilities measured for the ⁵¹Cr and ²⁴¹Am are presented in Tables 1 and 2, respectively, and compared with the literature. The total uncertainty in each value was obtained by quadrature, including all the uncertainties, namely: mass, decay, detector efficiency and statistics (one standard deviation). Table 3 gives the values of partial uncertainties involved in the emission probability determination.

For the 26.34 keV ²⁴¹Am gamma ray, the result of the present work corresponds to a value between lwahara et al. (2005) and Bé et al. (2004), and hence do not solve the disagreement between the two, suggesting the need of more measurements.

For the 320.08 keV ⁵¹Cr gamma ray, the value agrees with the literature values, confirming the value settled by Bé et al. (2004). For the KX-rays our values agree with all values presented within experimental uncertainties.

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