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Determination of ^{198}Au X-rays emission probabilities

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ABSTRACT

This work describes the measurements of the K X-ray and gamma-ray emission probabilities per decay of ^{198}Au performed at the Nuclear Metrology Laboratory (LMN) at the IPEN, São Paulo. The radioactive sample was obtained by means of ^{197}Au(n, γ)^{198}Au reaction irradiating an Au foil in a thermal neutron flux near the core of the IPEN 3.5 MW research reactor. The activity of samples was determined in a 4πβ–γ coincidence system, setting the gamma window at the 411.80 keV total energy absorption peak. The same samples were measured in two different spectrometers: a HPGe planar spectrometer with Be window, suitable for measurements in the low energy range and a coaxial REGe spectrometer. Both spectrometers were previously calibrated in a well defined geometry by means of standard sources calibrated in a 4πβ–γ coincidence system. MCNP4C Monte Carlo code was used for simulating the REGe spectrometer calibration curve, and a new version of code ESQUEMA was adopted for simulating the detection processes in the coincidence system, in order to predict the efficiency extrapolation curve.

1. Introduction

The radionuclide ^{198}Au decays with a half-life of 2.6950(7) d by β^- emission, populating the excited levels of ^{198}Hg, which emits K X-rays between 68.89 and 83.03 keV, a main gamma ray of 411.80 keV with emission probability of 95.5%, and two other gamma lines with 675.88 and 1087.69 keV with lower emission probabilities (IAEA, 2007).

The choice of this radionuclide is due to the need of new results in the low energy range, which are scarce in the literature. The most recent recommended values for the K X-ray emission probabilities (IAEA, 2007) are 0.00809(8) for 68.89 keV and 0.01372(12) for 70.81 keV. In this work, emission probabilities of these K X-rays and also 411.80 and 675.88 keV gamma-rays were experimentally obtained with absolute activity measurements and spectrometry methods.

Fig. 1 shows the ^{198}Au decay scheme. Due to this simple decay scheme, the 4πβ–γ coincidence method is most suitable for high accuracy activity determination.

In this work, we applied this method in two different ways and the results were compared: (1) measurements in a 4πβ–γ coincidence system applying the efficiency extrapolation technique; (2) simulation by Monte Carlo method of the interaction of β-particles and γ-rays with the detectors materials in order to predict the efficiency extrapolation curve and compare with experimental results.

To determine the X-rays emission probabilities, a careful study of the low energy spectra was necessary because in this region many multiplets appear which have to be resolved by deconvolution methods.

2. Methodology

2.1. Sample preparation

The sample was obtained by means of the ^{197}Au(n, γ)^{198}Au reaction in a thermal neutron flux near the core of the IPEN 3.5 MW research reactor, irradiating an Au foil by 5 min. After irradiation, the foils were dissolved into a warm mixed solution of HCl and HNO_3 in 3:1 ratio. A total of 12 samples were prepared by dropping known aliquots of the radioactive solution on 20 μg cm^{-2} thick collodion films, which were previously coated with 10 μg cm^{-2} gold. A seeding agent (CYASTAT SM) was used for improving the deposit uniformity and the sources were dried in a dissecator. The mass determination was performed using the pycnometer technique (Campion, 1975). No detectable impurities were found.

2.2. Activity determination by 4πβ–γ coincidence system

The activity of the ^{198}Au solution was determined by measuring the samples in a 4πβ–γ coincidence system and applying the efficiency extrapolation technique (Baerg, 1973). The system consisted of a gas-flow proportional counter with 4π

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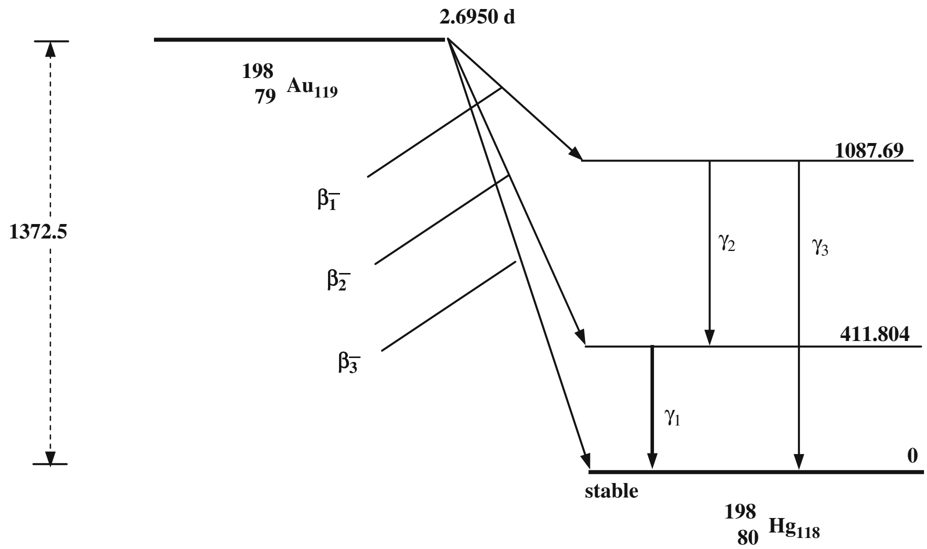

Fig. 1. Decay scheme of ^{198}Au. All energies are in keV.

geometry operating at +2050 V for detecting β-particles and electrons (β-channel) and a pair of 76 mm × 76 mm NaI(Tl) scintillation counters, positioned above and below the proportional counter, in order to detect γ-rays (γ-channel). The associated electronic system provides registration of β, γ and coincidence events by means of a time-to-amplitude-converter (TAC) coupled to a multichannel analyzer (Baccarelli, 2008). The proportional counter efficiency was around 97% and the acquisition time was 2000 s for each measurement.

The 411.80 keV total energy absorption peak was selected as the gamma window.

2.3. Monte Carlo simulation

The behavior of the apparent activity value of $N_\beta N_\gamma / N_C$ was simulated as a function of the proportional counter efficiency by Monte Carlo calculation using the ESQUEMA code (Takeda et al., 2005), which simulates the behavior of radiation decay and absorption in the 4πβ–γ system. The calculation was performed simulating the variation of proportional counter efficiency applying collodion absorbers above and below the radioactive sources. The number of histories was set to 1×10^7 events.

3. X-ray spectrometry

Two different spectrometers were used in this work: a planar HPGe for the low energy region, which is the main purpose of this work, and a REGe coaxial used to determine the emission probabilities per decay of ^{198}Au gamma-rays.

The emission probability was determined by means of the following equation:

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

where $S(E_{X,\ \gamma})$ is the counting rate under total absorption peak of energy $E_{X,\ \gamma}$, A is the absolute activity and $\varepsilon(E_{X,\ \gamma})$ is the detector efficiency to photons with energy $E_{X,\ \gamma}$. Corrections for background, cascade summing, dead time and self absorption were applied to the results. The cascade summing correction has been calculated by means of a Monte Carlo code developed at the LMN, which considers information from all ^{198}Au gamma-ray transitions (Dias et al., 2002).

The REGe coaxial detector with Be window with 500 μm thick yielded a 1.79 keV FWHM resolution at 1332.5 keV. The source to detector distance was 17.9 cm, and the efficiency calibration range was between 244 and 1408 keV using ^{60}Co, ^{133}Ba, ^{152}Eu and ^{166m}Ho standard sources, previously calibrated by the 4πβ–γ coincidence system. All samples were measured with acquisition times from 6×10^3 to 10^5 s, yielding a number of counts in the total energy absorption peak from 9×10^4 to 4×10^6. The spectra were analyzed by ALPINO code (Dias, 2001), which applies the method of simple integration of the peak.

The HPGe planar detector with 50 μm thick Be window showed an energy resolution of 160 eV (FWHM) at 5.9 keV. The source to detector distance was 10.4 cm, and the efficiency curve was obtained analyzing X-ray and gamma-ray spectra in the range from 5 to 80 keV with standards sources of ^{54}Mn, ^{55}Fe, ^{57}Co, ^{133}Ba, ^{152}Eu, ^{166m}Ho and ^{241}Am, calibrated by the 4πβ–γ coincidence system. Corrections for attenuation in Be window and air, as well as for germanium K-edge effects were applied, with theoretical calculation according to Debertin and Helmer (1988).

Seven samples were measured with acquisition times from 70×10^3 to 250×10^3 s, resulting in 10×10^3 to 150×10^3 to K X-rays counts at the 68.89 keV peak. Gamma-ray and X-ray spectra for efficiency curves and for emission probability determination were analyzed by means of COLEGRAM code (Ruellan et al., 1996). For gamma-ray peak fittings, a Gaussian distribution was adopted and a Voigt profile was utilized for X-rays peaks, fixing the Lorentzian width in accordance to the natural K X-rays lines width (Krause and Oliver, 1979). In both analyses, an exponential background was considered, after room background subtraction. The curve fitting to experimental data was performed by least square method.

4. Results and discussion

The extrapolation curve obtained by Monte Carlo for the activity determination in the coincidence system is presented in Fig. 2. In this figure the experimental values were normalized to Monte Carlo calculation at the same proportional counter efficiency.

The specific activity results for the ^{198}Au solution are presented in Table 1. The activity was calculated by code CONTAC developed at LMN (Dias, 2003) and the slope of the

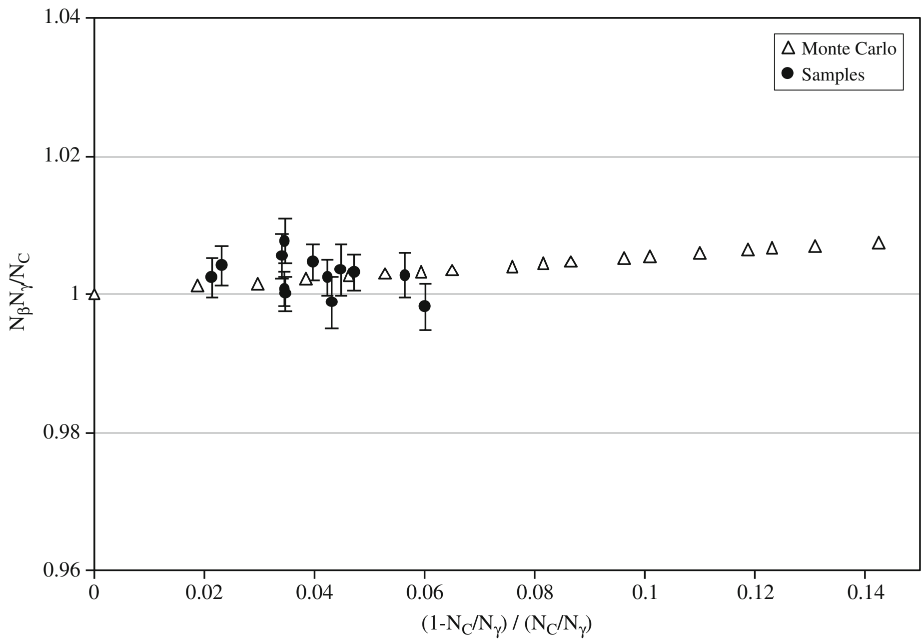

Fig. 2. Comparison of experimental data and Monte Carlo simulation of the extrapolation curve of $N_\beta N_\gamma/N_C$ as a function of inefficiency parameter.

Table 1
Comparison between different methods for activity determination.

Method	Activity (kBq g^{-1})	Uncertainty (%)
Experimental decay scheme correction	295.53	0.20
Simulation by Monte Carlo for collodion absorbers	294.92	0.13

The uncertainties correspond to one standard deviation.

Table 2
Uncertainty budget for ^{198}Au activity determination by 4πβ–γ coincidence method (one standard deviation).

Component	Uncertainty (%)
Counting statistics	0.06
Mass	0.10
N_C/N_γ parameter	0.11
Decay scheme correction factor	0.12
Total	0.20

extrapolation curve measured previously by the LMN (Dias, 1978), 0.04889(6), was applied. This activity value is compared with the Monte Carlo result and they are in agreement within the experimental uncertainties.

The uncertainty budget for this activity measurement is presented in Table 2. The main contributions come from decay scheme correction and N_C/N_γ parameter.

An example of processing of K X-ray region of ^{198}Au spectrum using code COLEGRAM can be seen in Fig. 3. The two peaks on the left are the $K_{\alpha2}$ and $K_{\alpha1}$, respectively, which can be separated with good accuracy. There are two triplets on the right side of the picture. The first corresponds to ($K_{\beta3}$+$K_{\beta1}$+$K_{\beta5}$) X-ray lines and the second one corresponds to ($K_{\beta2}$+$K_{\beta4}$+K_O) X-ray lines. The total emission intensity have been considered for each triplet.

Fig. 4 shows the efficiency curves for both detectors fitted by least square method using LOGFIT code (Dias and Moreira, 2005), in comparison with the experimental points. For the planar HPGe detector a sixth degree polynomial in log–log scale has been fitted. For the coaxial REGe detector a second degree polynomial in log–log scale has been fitted. The corresponding chi-square values were 1.12 and 1.18, respectively, indicating satisfactory fits. The dot line represents the Monte Carlo simulation with MCNP4C code applied to REGe spectrometer geometry which coincides with the polynomial fitting.

The final values of the emission probability of K X-rays and gamma-rays are presented in Table 3. These results were obtained by the weighted mean of the individual values from all samples, applying the covariance methodology (Smith, 1991). This table includes data from IAEA (2007), Hammed et al. (1992) and Chand et al. (1989), which are all in agreement within the experimental uncertainties, except for the 79.82–80.75 keV value from Chand et al. (1989) which is slightly higher.

The uncertainty budget is presented in Table 4. For most cases the predominant error comes from the HPGe detector efficiency. The high statistical error in the (82.42+83.03) keV X-ray peak is due to very low intensity (0.135%).

5. Conclusion

The extrapolation curve obtained by the 4πβ–γ coincidence method was compared with Monte Carlo prediction, using code ESQUEMA, and agreed well showing that this approach is reliable for predicting extrapolation curves.

The emission probabilities of several X-ray and two gamma-ray lines from ^{198}Au were measured absolutely and compared with the literature. The lack of experimental data for the emission probabilities per decay for this radionuclide obtained in the literature by absolute methods makes this comparison difficult. However, the existing values are in agreement within the experimental uncertainties, except for the 79.82–80.75 X-ray line taken from Chand et al. (1989) which is slightly higher.

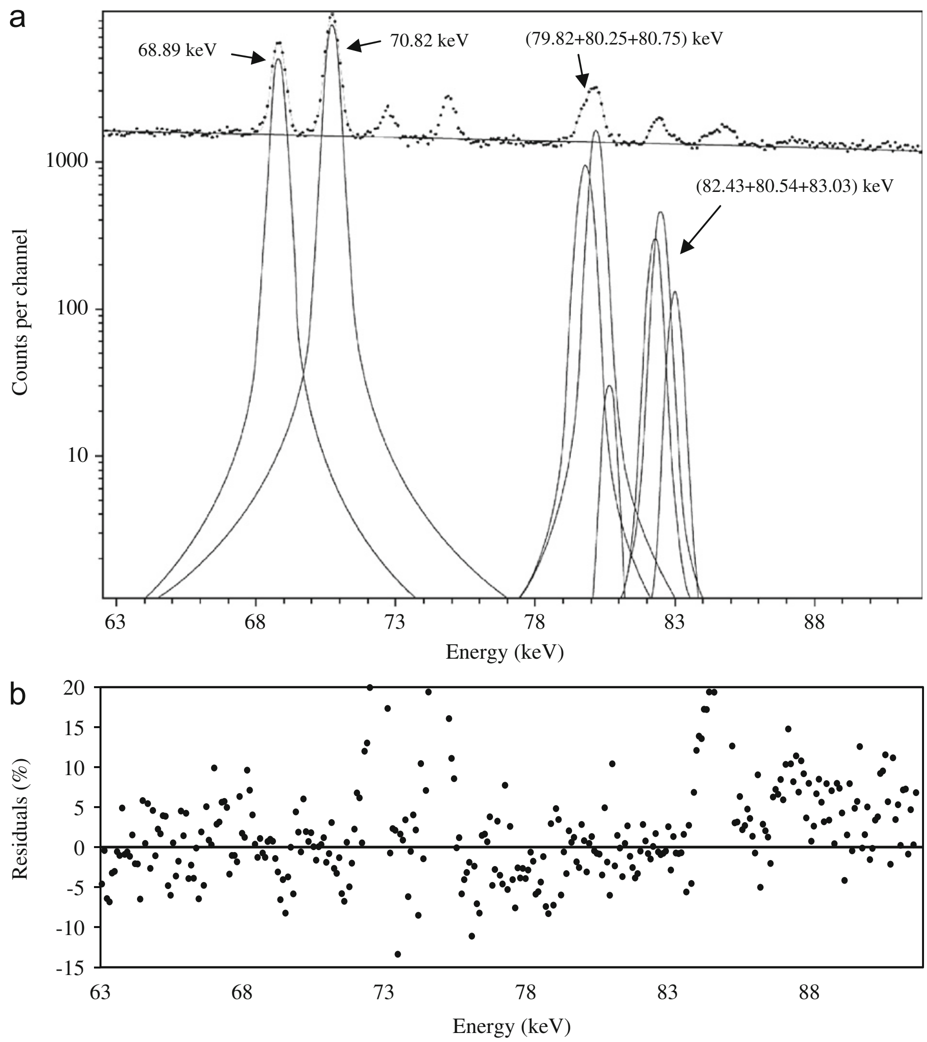

Fig. 3. Results of ^{198}Au spectrum in the *K* X-ray region, processed by code COLEGRAM (Ruellan et al., 1996). Part (a): the two peaks on the left are the $K_{\alpha 2}$ and $K_{\alpha 1}$, respectively. The two triplets on the right side of the picture correspond to ($K_{\beta 3}$+$K_{\beta 1}$+$K_{\beta 5}$) X-ray lines and the second one to ($K_{\beta 2}$+$K_{\beta 4}$+K_{O}) X-ray lines. Part (b): percent residues are presented as a function of the energy.

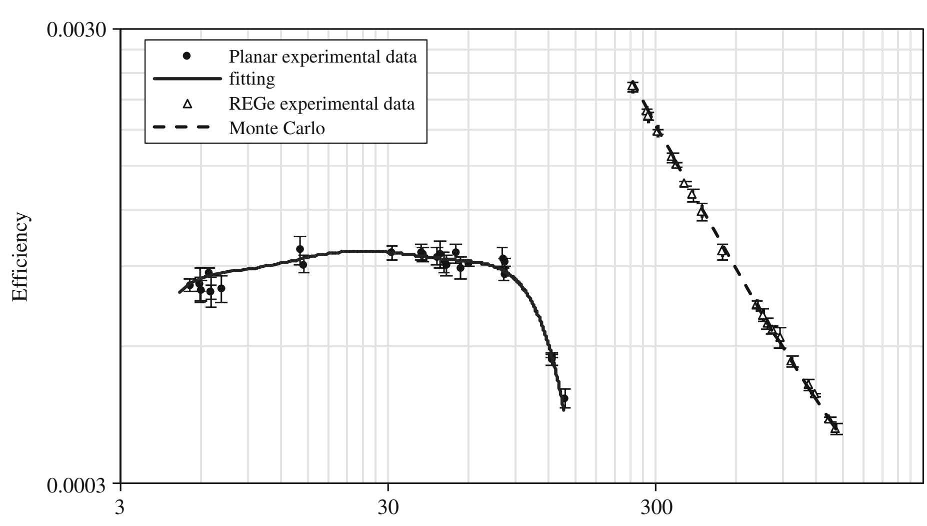

Fig. 4. Full energy peak efficiency curves for HPGe planar and REGe detectors.

Table 3
Emission probabilities per decay of K X-rays and gamma-ray of ^{198}Au, in comparison with the literature.

Energy (keV)	This work	IAEA (2007)	Hammed et al. (1992)	Chand et al. (1989)
68.89	0.00813(10)	0.00809(8)	–	0.00816(24)
70.82	0.01374(17)	0.01372(12)	–	0.0141(4)
79.82–80.75	0.00460(7)	0.00466(8)	–	0.00485(12)
82.43–83.03	0.00135(3)	0.00136(4)	–	0.00137(7)
411.80	0.9528(63)	0.9554(7)	0.9556(65)	–
675.88	0.00800(6)	0.00806(7)	0.00805(9)	–

Table 4
Uncertainty budget for ^{198}Au emission probability determination (one standard deviation).

Component	Uncertainty (%) (keV)					
	68.89	70.82	79.822–80.75	82.43–83.03	411.80	675.88
Counting statistics	0.66	0.44	0.25	1.63	0.10	0.49
Mass	0.10	0.10	0.10	0.10	0.10	0.10
Detector efficiency	1.07	1.12	1.45	1.55	0.56	0.48
Half-life	0.04	0.04	0.04	0.04	0.04	0.04
Dead time	0.27	0.24	0.28	0.26	0.26	0.28
Source activity	0.20	0.20	0.20	0.20	0.20	0.20
Total	1.30	1.24	1.51	2.27	0.66	0.77

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Discussion

Due to technical problems (unclear records) the transcription can not be published.