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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.

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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

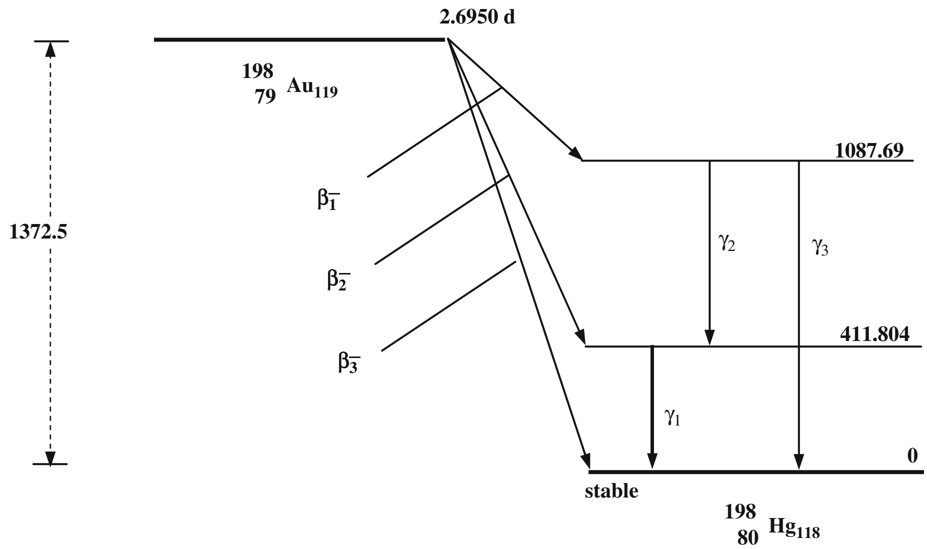

Fig. 1. Decay scheme of ^{198}Au. All energies are in keV.

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