Activity determination of a ^{201}Tl solution by 4πβ-γ and sum-peak coincidence methods

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Abstract: ^{201}Tl is used in nuclear medicine in cardiac imaging for evaluating the injury level in cardiac muscle at rest and exercise. In this work the activity concentration of a ^{201}Tl radioactive solution has been absolutely determined using the 4πβ-γ coincidence and sum-peak coincidence methods. The presence of ^{202}Tl radioactive impurity that imposes some difficult in the activity measurements was taken into account in the measurements. In the sum-peak method a planar germanium detector was used. The half-lives were evaluated by the reference source method and the results obtained were (3.033 ± 0.004) d and (12.320 ± 0.163) d, respectively, for ^{201}Tl and ^{202}Tl.

Keywords: ^{201}Tl, activity, nuclear medicine, coincidence counting, sum-peak counting.

1. INTRODUCTION

The determination of the activity or disintegration rate of radionuclides and the nuclear decay parameters such as half-lives and photon emission intensities, mainly for those used in nuclear medicine, are necessary to allow the correct calibration of the measuring instruments and consequently, the optimization of the internal dose delivered to the patients.

The principal method used for activity standardization of radionuclides by most National Metrology Institutes worldwide is the primary 4πβ-γ coincidence counting method. Another method currently used is the sum-peak coincidence, mainly for those radionuclides which decay by electron capture with short half-lives. The sum-peak method is a primary measurement technique that combines gamma-ray spectrometry and sum coincidence counting, requiring only one photon detector with associated electronics. The activity is determined from the count rates of the full absorption peaks of a single X- and gamma-rays, the sum-peak count rate of photons in true coincidence and the total count rate of the spectrum.

The half-life was determined by the reference source method which uses the simultaneous

measurements of gamma spectrometry of the sample and the reference in a germanium detector. This feature allows optimizing the analysis of time and eliminating the influence of instrumental and environmental interferences. Unlike other methods such as utilizing the ionization chamber, it does not depend on the device stability, and in the particular case, not depends on the purity of the sample.

The half-life is an important nuclear parameter in identifying and quantifying a radionuclide, so there is a demand for research to determine more accurate half-lives of radionuclides used in calibration of equipment used to measure gamma and X-rays emitters in nuclear medicine, radiation protection and dosimetry, environmental monitoring, nuclear security programs, reactors and treatment of radioactive waste.

This work describes the standardization of a ^{201}Tl solution by two primary methods, the quantification of ^{202}Tl impurity by gamma spectrometry and the determination of half-lives of ^{201}Tl and ^{202}Tl by the reference source method.

2. METHODOLOGY

2.1. 4πβ-γ coincidence counting method

There is an abundant literature on the application of 4πβ-γ coincidence method for standardization of radioactive sources. It is the primary method that has great versatility as may be used for those radionuclides which decay by alpha-gamma and beta-gamma emission, electron capture and pure beta emitters [1,2].The 4πβ-γ coincidence system used in this work consists of a gas flow 4π proportional counter coupled to a 10.2 cm ×10.2 cm crystal of NaI(Tl). The counting gas of the 4π counter is a mixture of 90% argon and 10% methane operating at 0.1 MPa pressure. Characteristics X-rays and electron Auger originating from ^{201}Tl are counted in the proportional counter and the gamma counts were confined to a window covering the 167 keV line in the scintillator crystal. In order to avoid the interference effects of the sum-coincidences of X-rays originating from electron capture events (69 to 83 keV) in the gamma counting, a cadmium absorber of 2 mm thick was placed between the source and the scintillator of NaI (Tl). To obtain the activity it is applied the method of efficiency extrapolation. In this method the plot of $(N_\beta N_\gamma)/N_c$ as a function of $(1-N_c/N_\gamma/\ N_c/N_\gamma)$ gives the source activity N_0 when $N_c/N_\gamma = \varepsilon_{X,A} \rightarrow 1$. Here $\varepsilon_{X,A}$ is the count efficiency of proportional counter for X-rays and electron Auger and N_β, N_γ and N_c are β, γ and coincidence count rates, respectively. The variation of efficiency is carried out using aluminized mylar absorbers of various thicknesses on the counts of the proportional counter originating from the electron capture events. In this work we use the expression developed by Funck [3]

$$\frac{N_\beta N_\gamma}{N_c} = N_0\left(1 + c_1 + c_2\frac{1-\varepsilon_{X,A}}{\varepsilon_{X,A}}\right) \qquad (1)$$

where c_1 and c_2 are constants depending on decay scheme data.

2.2. Sum-peak coincidence method

It is a primary method of measuring the activity being determined from the total count rate of photon spectrum, the sum-peak count and emission of X- and gamma-rays, and the relationship between them given by equation 2 [4].

$$N_0 = N_T + \frac{N_X N_\gamma}{N_{X\gamma}} - \frac{N_{XX}N_\gamma^2}{N_{X\gamma}^2} \qquad (2)$$

where N_0 is the activity, N_T is the spectrum total count, N_X is X-ray count, N_γ is the gamma count, $N_{X\gamma}$ is the sum count of X- and gamma-rays and N_{XX} is the sum count of X-rays.

2.3. Reference source method for half-life measurements

The reference source method is based on the ratio R_t of the counts of the radionuclide to be measured and the standard, according to equation 3 [5].

$$R_t = \frac{C_t}{P_t} \quad (3)$$

C_t is the count of the radionuclide to be measured in time t and P_t is the count of the reference at the same time t. Based on the radioactive decay law, the equation 3 can be rewritten as

$$R_t = \frac{C_0 e^{-\lambda_C t}}{P_0 e^{-\lambda_P t}} = \frac{C_0}{P_0} e^{-(\lambda_C - \lambda_P)t} = R_0 e^{-\left(\frac{ln2}{T_C} - \frac{ln2}{T_P}\right)t} \quad (4)$$

where C_0 and P_0 are the gamma ray count of the radionuclide to be measured and the reference in time t=0, respectively, λ_C and λ_P are decay constants of the radionuclide to be measured and the reference, respectively, T_C is the half-life of radionuclide to be measured and T_P is the reference half-life.

The equation can be rearranged by

$$ln\frac{R_t}{R_0} = -\left(\frac{ln2}{T_C} - \frac{ln2}{T_P}\right)t \quad (5)$$

and

$$T_C = \frac{ln2}{\frac{ln2}{T_P} - \left(\frac{lnR_t}{t} - \frac{lnR_0}{t}\right)} \quad (6)$$

where $\ln(R_t/t) - \ln(R_0/t)$ is the slope *a* of the plot $\ln(R_T)$ as function of time t. The final equation can be rearranged as

$$\boldsymbol{T_C = \frac{1}{\frac{1}{T_P} - \frac{a_1}{ln2}}} \quad (7)$$

3. RESULTS AND DISCUSSION

3.1. Activity determination

Five sources for the determination of the activity by 4πβ-γ coincidence counting method were prepared from a solution of ^{201}Tl provided by IPEN/SP. One or two drops of the solution (10 to 20 mg) were deposited on thin VYNS films (copolymer of vinyl chloride and vinyl acetate), approximately 15 μg/cm^2 thick, coated with a gold layer of 20 μg/cm^2 thick on both sides of the film. The extrapolated value for determining the activity must be multiplied by a correction factor due to non-detection of low energy X-rays and Auger electrons [3]. This factor depends upon the nuclear decay scheme parameters such as conversion coefficients, abundance ratios, capture probability and fluorescence yields and for ^{201}Tl is estimated to be 0.9908 for the counting system conditions used in this work (Equation 1). Besides, the radiochemical impurities of ^{200}Tl and ^{202}Tl, detected by gamma spectrometry, should be subtracted. The percentage of impurity was 1.15% and 0.01% of ^{202}Tl and ^{200}Tl, respectively, relative to ^{201}Tl in this exercise in the reference date. From the weighted average of five activity results obtained, the value of (1898.46 ± 13.67) kBq/g at the reference date of May 01, 2015, 12h00 was evaluated. A typical extrapolation curve of one source is showed in Fig 1.

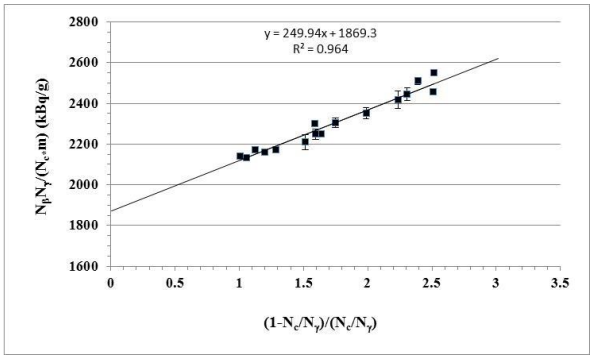

Figure 1: Typical extrapolation curve of experimental data for coincidence measurements of one source of ^{201}Tl. The extrapolated value for $(1\text{-}N_c/N_\gamma)/(N_c/N_\gamma)$ gives the activity concentration of the source.

The uncertainty components for activity determination of ^{201}Tl by the 4πβ-γ coincidence method is presented in Table 1.

Table 1: Uncertainty components for activity determination of ^{201}Tl by coincidence method.

Uncertainty component due to	Relative standard uncertainty (%)
Counting statistic (included on fitting procedure	0.90
Fitting procedure	0.60
Weighing	0.05
Dead time	0.13
Resolving time	0.09
Delay mismatch between gamma and beta pulses	0.34
Background	0.02
Decay (due to uncertainty on ^{201}Tl half-life)	0.11
Combined standard uncertainty	0.72
Expanded uncertainty (k=2)	1.4

For applying the sum-peak coincidence method for activity determination, one point source of ^{201}Tl on polystyrene film support was submitted to two measurement cycles with the source placed on the top of the high purity planar germanium detector. The real counting time was 3600 seconds with dead time of 56 and 35%, respectively. The time interval between the cycles was approximately 3 days. The activity concentration was determined using the equation 2, section 2.2, giving 1751.24 kBq/g for ^{201}Tl and 16.708 kBq/g for ^{202}Tl. The same source was resubmitted to 2 measurement cycles in the germanium detector, but with the dead time of 19 and 7%, respectively, real counting time of 3600 s and the time interval between two cycles of approximately 3 days. In this case, the activity concentration was 1887.99 kBq/g for ^{201}Tl and 22.218 kBq/g for ^{202}Tl. The latter result is consistent with the coincidence method showing the importance of the system dead time in the activity calculation by sum-peak method.

The statistical uncertainty (0.28%) in the count is the component of type A; type B components are weighing (0.05%), decay scheme parameters (0.50%), the half-life (0.31%) and the background radiation (0.22%) given total standard uncertainty of 0.69%. The photon spectrum of a ^{201}Tl source obtained by planar germanium detector for application of sum-peak method is showed in Fig. 2. In this spectrum one can observe the photopeaks of characteristics K_α and K_β X-rays (68.90 to 80.7 keV), the coincidence sum-peak of (K_α+K_β X-rays, the 167 keV γ-ray and the coincidence sum-peak of (K_α+167) keV γ–ray and (K_β +167) keV γ–ray.

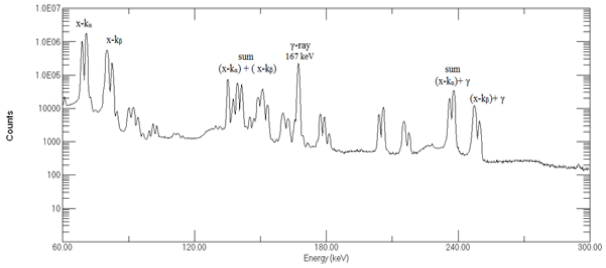

Figure 2: Photon spectrum of a ^{201}Tl source.

The gamma spectrometry with a coaxial germanium detector of 50% efficiency, aluminum window of 1.00 mm thick and energy resolution of 0.90 keV and 1.90 keV for 122.06 keV of ^{57}Co and 1332.5 keV of ^{60}Co, respectively, was used to validate the results so far obtained. An efficiency curve as a function of energy (ε versus E) was determined using standard point sources of various photon-emitting radionuclides with energies ranging from 40 to 2500 keV. A second order hyperbola $Ln(\varepsilon)=A+B/(LnE)+C/(LnE)^2$ was fitted to the experimental data using the Levenberg-Marquardt algorithm based on the curve fitting least square method [6]. The fitting parameters calculated were A = -15.966 ± 0.221, B = 97.590 ± 2.595 ± and C = -239.385 ± 7.547, degree of freedom 74 and Chi square 13.895. The critical Chi square is 95.081 for α = 0.05. The photopeak efficiency for 167 keV gamma ray of ^{201}Tl, 440

keV for ^{202}Tl and 368 keV for ^{200}Tl are calculated by interpolating ε versus E curve for these energies. The interpolated efficiencies from the fitted curve are presented in Table 2.

Table 2. Efficiencies obtained by interpolation of experimental efficiency versus energy calibration curve.

Energy (keV)	ε	Standard relative uncertainty of ε (%)
167 (^{201}Tl)	0.002389	018
368 (^{202}Tl)	0.001825	0.25
440 (^{200}Tl)	0.001671	0.23

The activity is determined by using the equation below:

$$A = \frac{C}{\epsilon * I_\gamma} \quad (8)$$

where A is the activity (Bq), C is the count rate of photopeak of energy E (count/s) and I_γ is the gamma intensity of energy E. Fig. 3 shows the ε versus E calibration curve of germanium detector.

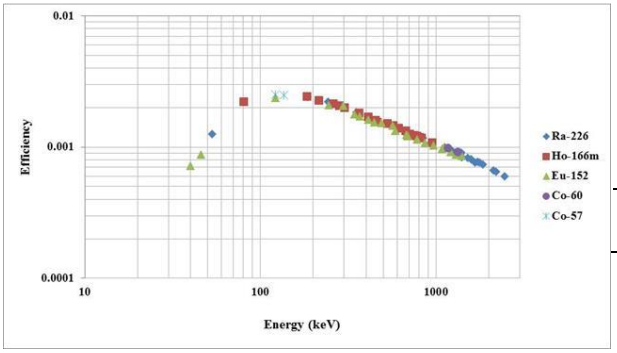

Figure 3: Efficiency versus energy calibration curve of germanium detector.

Three point sources of ^{201}Tl prepared in the polystyrene film holder were measured at 10 cm from the top of the germanium detector. The count times were 74000 s (4.80% of dead time), 78000 s (5.17% of dead time) and 86000 s (1.91% of dead time). The results are shown in Table 3. Table 4 presents the results of radiochemical impurities of the ^{200}Tl and ^{202}Tl.

Table 3: Activity concentration results of ^{201}Tl obtained from de efficiency curve method.

Source	Activity concentration (kBq/g)	Standard uncertainty (kBq/g)
01	1883.64	12.82 (0.68%)
02	1883.88	12.74 (0.68%)
03	1882.58	12.50 (0.66%)
Weighted mean	1883.36	7.32 (0.39%)

Table 4: Radiochemical impurities detected in the ^{201}Tl solution.

Source	Activity concentration (kBq/g)		Standard uncertainty (kBq/g)	
	^{202}Tl	^{200}Tl	^{202}Tl	^{200}Tl
01	21.69	0.100	0.156 (0.72%)	0.002 (1.9%)
02	21.56	0.100	0.142 (0.66%)	0.002 (1.8%)
03	21.17	0.083	0.112 (0.53%)	0.004 (4.6%)
Weighted mean	21.40	0.098	0.229 (1.1%)	0.005 (5.5%)

3.2. Half-life determination

The half-life of ^{201}Tl was determined using the methodology of source reference described in section 2.3. The reference sources used were ^{137}C and ^{54}Mn. A mixed point source of ^{201}Tl + ^{137}Cs + ^{54}Mn in polystyrene film support was subjected to two counting cycles in the germanium

detector. The spectral inspection revealed the presence of gamma emitting impurities of ^{202}Tl (440 keV) as can be seen in Fig. 4.

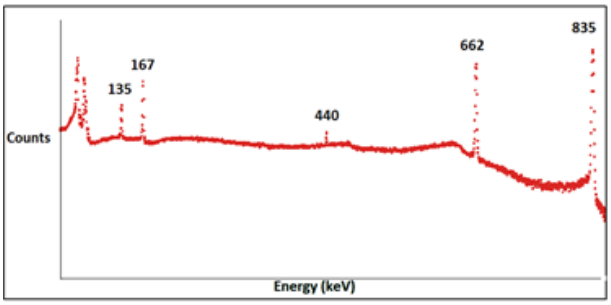

Figure 4: Gamma spectrum of ^{201}Tl+^{137}Cs+^{54}Mn point source by germanium detector.

In the first cycle 40 spectra were obtained with dead time ranging from 6.8 to 6.6%. Measurements of the second cycle performed a week later, originated 50 spectra obtained with dead time ranging from 5.4 to 5.3%. The total counting time, including the two cycles, corresponded to approximately 4 half-lives. Table 4 shows the values of the half-lives determined for each one of the reasons, emphasizing each gamma line and each reference. The values found show that the use of the gamma line of 135.34 keV (gamma intensity P_γ of 2.56%), for both reference, showed a large difference from the value determined by the gamma line of 167 keV (P_γ =10.2), the latter being consistent with the values found in the literature. Using the same spectra used to determine the half-life of ^{201}Tl, the half-life of the radiochemical impurity ^{202}Tl was calculated yielding a value of (12.320±0.163) d. Table 5 shows the results of half-lives of ^{201}Tl evaluated according equation 7, section 2.3.

Table 5: Half-lives of ^{201}Tl evaluated by source reference method.

Gamma energy (keV)	Reference	Half-life (d)
135.34	^{137}Cs 661.66 keV	2.928 ± 0.009 (0.32%)
	^{54}Mn 834.84 keV	2.929 ± 0.009 (0.32%)
167.41	^{137}Cs 661.66 keV	3.033 ± 0.009 (0.14%)
	^{54}Mn 834.84 keV	3.033 ± 0.004 (0.14%)

For comparison, the half-lives of ^{201}Tl and ^{202}Tl found in the literature [7] are respectively, 3.043±0.003 and 12:23±0.02 d.

4. CONCLUSIONS

The results obtained show that the coincidence sum-peak method can be used as an alternative to the primary standardization of radionuclides. Some specific conditions are required for the implementation of the method, such as the time interval between measurement cycles, the activity of the solution since the measurements with high dead times showed large discrepant results regarding the 4πβ-γ coincidence method. The results also showed that the method of the source reference associated with gamma spectrometry techniques allows determining the half-life of the radionuclide either as the radiochemical impurity using the same measurement, despite the very low concentration of the impurity relative to the main radionuclide.

7. REFERENCES

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