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Monte Carlo simulation to positron emitter standardized by means of 4πβ-γ coincidence system—Application to ^{22}Na

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ABSTRACT

The present work describes the methodology for predicting the behavior of extrapolation curves obtained in radionuclide standardization by 4πβ-γ coincidence measurements, applied to ^{22}Na, developed at the Laboratório de Metrologia Nuclear of IPEN-CNEN/SP (LMN–Nuclear Metrology Laboratory). The LMN system consists of a proportional counter (PC) in 4π geometry coupled to a single or a pair of NaI(Tl) scintillation crystals. Two standardization techniques were used: the Sum-Peak and the Nuclear-Peak methods. The theoretical response functions of each detector have been calculated using the MCNPX Monte Carlo code. The code ESQUEMA, developed at LMN, has been used for calculating the extrapolation curve in the 4πβ-γ coincidence experiment. Modifications were performed in order to include response tables for positrons and coincidences with annihilation photons. From the calibration results it was possible to extract both the activity value and the positron emission probability per decay. The latter was compared with results from the literature.

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1. Introduction

The Nuclear Metrology Laboratory (LMN) at IPEN, in São Paulo, Brazil, has developed a methodology for predicting the behavior of extrapolation curves obtained in radionuclide standardization by 4πβ-γ coincidence measurements. In previous papers this methodology has been applied to pure beta (Dias et al., 2006) and beta-gamma (Takeda et al., 2005) as well as electron capture-gamma emitting radionuclides (Dias et al., 2006).

The present work describes this application to ^{22}Na, an important standard in radionuclide metrology. This radionuclide decays by β^+ (89.892(17) %) and Electron Capture (10.11(11) %) with a half-life of 2.6027(10) y, mainly to the 1275 keV excited state of ^{22}Ne, as shown in Fig. 1 (Table de Radionucléides, 2006). The measurements were carried out by LMN during an international comparison coordinated by the PTB and mediated by the BIPM, in 1973 (Moura and Nuevo Jr., 1973). At that time the results were used only for obtaining the activity of the radioactive solution by means of two 4πβ(PC)-γ techniques, namely: the Sum-Peak and the Nuclear-Peak methods. The positron emission probability per decay was taken from the literature and applied to the activity results.

In the present approach, these data have been re-analyzed applying Monte Carlo modeling by means of the code ESQUEMA (Takeda et al., 2005; Dias et al., 2006) which has been modified for β^+-gamma emitters. New theoretical extrapolation curves were calculated and applied to the data. From these results the positron emission probability was derived, together with the radioactive solution activity.

2. Methodology

2.1. Coincidence equations

Full descriptions of the coincidence equations applied to ^{22}Na can be found elsewhere (Williams, 1964; Nähle et al., 2008). The formalism described below was derived for the specific conditions of the LMN calibration. The Auger electrons have energies below 1 keV and were assumed to be cutoff by thin absorbers used above and below the radioactive sources.

2.2. Sum-Peak method

In this case, the gamma-ray window was set to include only the total absorption energy window of the (511+1275=1786) keV Sum-Peak. In this condition, the counting rates in the beta, gamma and coincidence channels are reduced to (Nähle et al., 2008):

$$N_\beta = N_0[h\varepsilon_\beta + p_{\beta01}(1-\varepsilon_\beta)\varepsilon'_{\beta\gamma} + p_{EC01}\varepsilon_{\beta\gamma}] \quad (1)$$

$$N_\gamma = N_0[p_{\beta01}\varepsilon_\gamma(1786)] \quad (2)$$

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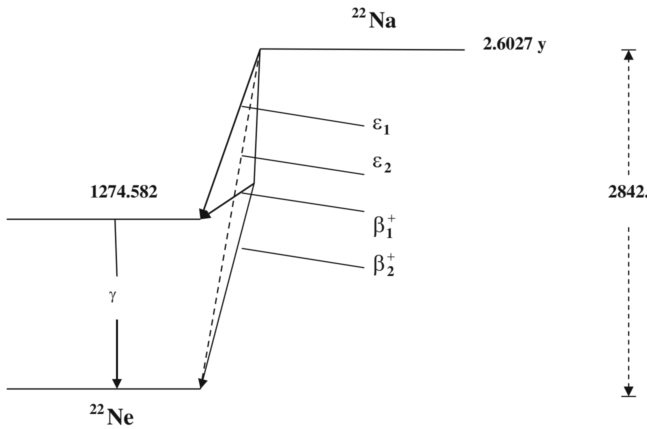

Fig. 1. Decay scheme of ^{22}Na (Table de Radionucléides, 2006).

$$N_C = N_0\{p_{\beta01}[\varepsilon_\beta + (1-\varepsilon_\beta)\varepsilon'_{\beta\gamma}]\varepsilon_\gamma(1786)\} \tag{3}$$

Therefore:

$$\frac{N_\beta N_\gamma}{N_C} = N_0 \frac{\left[h + p_{\beta01}\dfrac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\varepsilon'_{\beta\gamma} + \dfrac{p_{EC01}\varepsilon_{\beta\gamma}}{\varepsilon_\beta}\right]}{1 + \dfrac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\varepsilon'_{\beta\gamma}} \tag{4}$$

where N_0 is the source disintegration rate; h is the total β^+ transition probability; $p_{\beta01}$ is the β^+ transition probability to the excited state; p_{EC01} is the EC transition probability to the excited state; ε_β is the β^+ detection probability of the 4π(PC); $\varepsilon_{\beta\gamma}$ is the gamma-ray detection probability of the 4π(PC); ε_γ is the gamma-ray detection probability; $\varepsilon_{\beta\gamma}{}'$ is the gamma-ray detection probability of at least one out of the three gamma-rays (1275 and 511 keV) from the β^+ transition in the 4π(PC) (Nähle et al., 2008).

In the extrapolation limit where $(1-\varepsilon_\beta)\rightarrow 0$, if the activity N_0 is known, h can be determined. The value of N_0 has been obtained by the Nuclear-Peak method, as described below.

2.3. Nuclear-Peak method

In this case, the gamma-ray window was set to include only the total absorption energy peak of the 1275 keV gamma-ray. In this condition, the count rates in the beta, gamma and coincidence channels become (Nähle et al., 2008):

$$N_\beta = N_0[h\varepsilon_\beta + p_{\beta01}(1-\varepsilon_\beta)\varepsilon'_{\beta\gamma} + p_{EC01}\varepsilon_{\beta\gamma}] \tag{5}$$

$$N_\gamma = N_0[p_{\beta01}\varepsilon_\gamma(1275,\beta) + p_{EC01}\varepsilon_\gamma(1275,EC)] \tag{6}$$

$$N_C = N_0\{p_{\beta01}[\varepsilon_\beta + (1-\varepsilon_\beta)\varepsilon'_{\beta\gamma}]\varepsilon_\gamma(1275) + p_{EC01}\varepsilon_\gamma(1275)\varepsilon_{\beta\gamma}\} \tag{7}$$

Therefore:

$$\frac{N_\beta N_\gamma}{N_C} = N_0 \frac{\left[h + p_{\beta01}\dfrac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\varepsilon'_{\beta\gamma} + \dfrac{p_{EC01}\varepsilon_{\beta\gamma}}{\varepsilon_\beta}\right]}{p_{\beta01}\left[1 + \dfrac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\varepsilon'_{\beta\gamma}\right] + \dfrac{p_{EC01}\varepsilon_{\beta\gamma}}{\varepsilon_\beta}} \tag{8}$$

In the extrapolation limit where $(1-\varepsilon_\beta)\rightarrow 0$, N_0 can be determined.

2.4. Standardization setup

The $4\pi\beta$-γ coincidence system used in this calibration consisted of a proportional counter (PC) in 4π geometry coupled to a single or a pair of NaI(Tl) scintillation crystals. The coincidence technique was performed by measuring the positrons in the proportional counter. Since the Auger-electron energies were below 1 keV, these particles were cutoff by placing thin absorbers below and above the radioactive sources.

For the Sum-Peak method the system has a pair of NaI(Tl) crystals in order to increase the detection efficiency. The beta efficiency was changed by applying absorbers below and above the sources and the (511+1275) keV full energy absorption peak was selected for the gamma gate. For the Nuclear-Peak method the full energy absorption peak corresponding to 1275 keV gamma-rays was selected for the scintillation counter window. This system has only a single NaI(Tl) scintillation counter. In this case, the gamma-ray efficiency was changed by moving this NaI(Tl) counter gradually away from the PC counter, plotting the observed activity as a function of the gamma-ray efficiency and extrapolating to zero efficiency.

Although Eq. (8) does not depend explicitly on the gamma-ray efficiency, there is a contribution from the Compton pulses originating from the summing events which are located below the 1275 keV total absorption peak. The gamma-ray efficiency changes with $1/r^2$ (r being the distance between the NaI crystal and the radioactive source) whereas the summing events change with $1/r^4$, because they come from the product of efficiencies. Therefore, the summing events diminish as the distance increases, going to zero at infinite distance.

2.5. Monte Carlo simulation

The theoretical response functions of each detector have been calculated using MCNPX (ORNL, 2006) Monte Carlo code. The validation of these functions was performed in previous papers by successfully applying the simulation to other beta-gamma and EC-gamma nuclides (Dias et al., 2006; Takeda et al., 2005). The use of MCNPX was introduced in order to provide calculation for positrons as the radiation source and updated data libraries contained in the earlier version MCNP-4C. A full description of the $4\pi\beta$-γ system was developed including details of source substrate and absorbers. Two geometrical models were created: one for the Sum-Peak method and another one for the Nuclear-Peak method, because of the different number of NaI detectors involved in each of them. An energy range from 1.4 keV up to 4 MeV has been selected for positrons in the 4π detector selecting a total of 298 energy bins in a stepwise scale. For gamma-rays, the energy range was from 48 keV to 3.0 MeV in 752 uniform energy bins. Additional calculations were performed to obtain response functions for annihilation photons detected in the NaI(Tl), in different radioactive source absorber conditions and different distances between the NaI detector and the 4π(PC) counter. The numbers of histories followed were 5×10^4 for positrons and 2×10^6 for gamma-rays.

The code ESQUEMA (Takeda et al., 2005), developed at LMN, has been used for calculating the extrapolation curve in the $4\pi\beta$-γ coincidence experiment. In this way, Eqs. (4) and (8) could be reproduced theoretically as a function of the beta efficiency. Modifications were performed in the code to include response tables for positrons and coincidences with annihilation photons. For this purpose the decay scheme has been modified in order to include explicitly annihilation photon contribution, as shown in Fig. 2. In this figure, the first part of decay scheme corresponds to the positron-gamma decay branch. The photon emission produced by positron annihilation is indicated as $\gamma_\pm$ transition. The second part corresponds to the EC decay branch. The Fermi function was also modified in the code for positrons in accordance with the literature (Evans, 1955).

One difficulty in simulating positron emission resides in the annihilation process. When there is no absorber around the

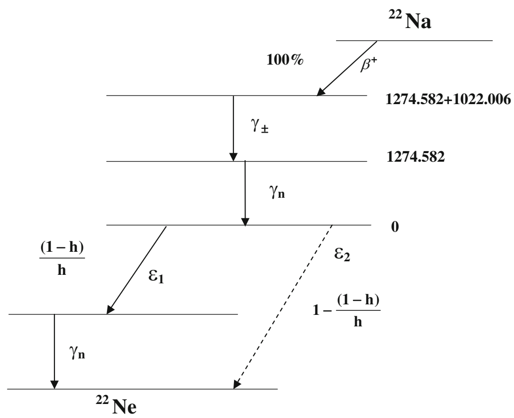

Fig. 2. Alternative decay scheme of ^{22}Na developed for the Monte Carlo simulation. The photon emission produced by positron annihilation is indicated as $\gamma_{\pm}$ transition. Parameter h is the total β^+ transition probability.

radioactive source, the positron tends to annihilate outside the source, mainly in the gas or inside the wall of the proportional counter. However, the presence of absorbers makes the annihilation occur near the radioactive source or between the source and the absorber. This effect changes the gamma-ray efficiency for annihilation photons in the NaI detector. The code ESQUEMA changes the beta efficiency by applying absorbers above and below the radioactive source. Therefore, there will be a dependence of the gamma-ray efficiency in the NaI detector with the positron energy sorted from the Fermi function and with the absorber thickness. These effects were evaluated by calculating the NaI response functions for annihilation photons, by means of the code MCNPX, for several positron energies and absorber configurations.

A functional relationship between positron energy and total gamma-ray efficiency in the NaI detector for annihilation photons has been developed as follows:

$$\varepsilon_T = a_0 + a_1 \frac{1}{(1+\exp z)} \tag{9}$$

where

$$z = \frac{\ln(E_0) - \ln(E_1)}{2.3548\,FWHM} \tag{10}$$

$\boldsymbol{a}_0$ and $\boldsymbol{a}_1$ are the asymptotic total gamma-ray efficiency values for low energy (1.4 keV) and high energy (1 MeV), respectively; $\boldsymbol{E}_0$ and $\boldsymbol{E}_1$ are the positron energies at the low energy region (1.4 keV) and after emerging the radioactive source absorber, respectively.

Eq. (9) was used by the code ESQUEMA in place of the total gamma-ray efficiency at NaI calculated by MCNPX at the source position, in order to simulate more accurately the effect of positron annihilation outside the radioactive source. In addition, an effective $\boldsymbol{a}_0$ value was adopted in order to account for the correlation between annihilation photons coming from the same positron, and thus emitted in opposite directions. This effect is not taken into account in the present version of ESQUEMA, and enhances the detection probability of these annihilation photons, as the two NaI detectors are situated on opposite sides with respect to the 4π proportional counter. The effective $\boldsymbol{a}_0$ value was established by verifying the experimental gamma-ray efficiency value at the 511 keV total energy absorption peak.

Fig. 2 shows the alternative ^{22}Na decay scheme used for the Monte Carlo simulation. Two decays were included: the first corresponds to the β^+ transition and the positron annihilation photon emission is indicated by the 1022 keV transition. For this transition, the differential pulse height spectrum of the NaI detector was calculated by MCNPX code at the average positron energy equal to 215.6 keV and included in the response table in place of the usual gamma-ray spectrum produced by the NaI detector at 1022 keV energy. The second decay corresponds to the EC transition. The intensities of EC branches ε_1 and ε_2 were set to give the correct branching ratios when included with the β^+ transition.

3. Results and discussion

Fig. 3 shows the variation of the NaI total gamma-ray efficiency produced by annihilation, as a function of the positron energy, calculated by the code MCNPX. The values were calculated without external absorbers around the radioactive source. The behavior follows approximately the shape given by Eq. (9). The final values of $\boldsymbol{a}_0$ and $\boldsymbol{a}_1$ were 0.34 and 0.07, respectively, using two NaI crystals close to the 4π proportional counter. These values were determined by applying a least squares fit to the experimental points shown in Fig. 3. The behavior of this shape can be explained by considering that the positron annihilation occurs close to the radioactive source at low positron energies and far from the source at high positron energies. Close to the source the geometrical gamma-ray efficiency is lower due to smaller solid angle. Far from the source, many positrons annihilate closer to the NaI crystal, increasing the gamma-ray efficiency.

Fig. 4 shows the experimental extrapolation curve obtained by means of the Sum-Peak method, changing the absorber thickness above and below the radioactive source. The experimental points have an approximately linear trend with positive slope caused by variation in the NaI gamma-ray efficiency for annihilation photons produced at different distances from the radioactive source. The Monte Carlo calculation agrees well with the experimental points. Running the code ESQUEMA for 8×10^6 events, the statistical uncertainty per data point in the calculation ranges from 0.01% to 0.16%, and the theoretical behavior is approximately linear. The behavior of experimental points for source No. 2 agrees quite well with the Monte Carlo simulation. The fluctuations in the experimental points for source No. 1 can be attributed to small instabilities of the coincidence system during data collection.

Table 1 presents the partial uncertainties involved in activity determination and in the parameter $\boldsymbol{h}$ (total β^+ transition probability). As can be seen, the parameter $(\mathbf{1}-\boldsymbol{h})/\boldsymbol{h}$ (EC/β^+

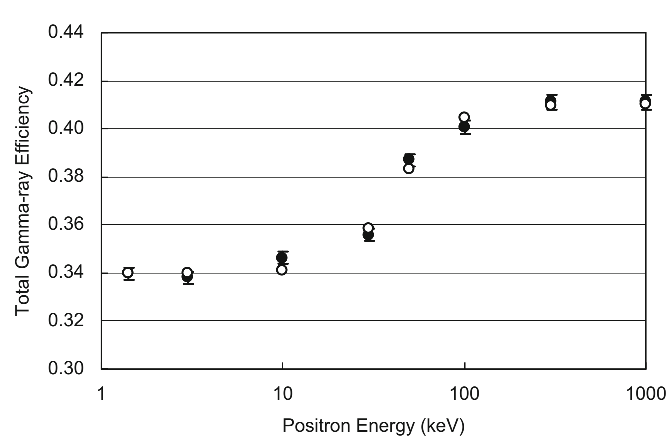

Fig. 3. NaI total gamma-ray efficiency for annihilation process, as a function of the positron energy. The behavior follows approximately Eq. (9). The closed markers are experimental values and the open dots are fitted values.

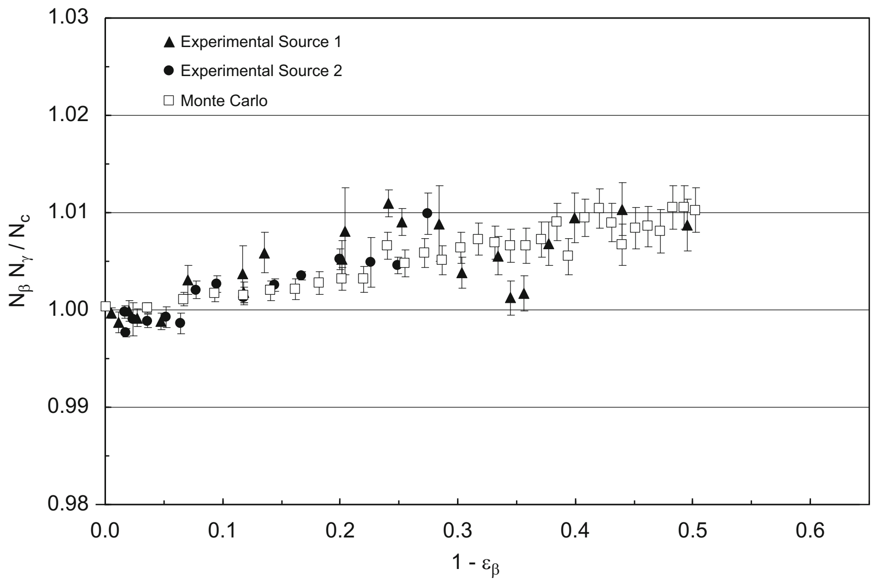

Fig. 4. Extrapolation curve for ^{22}Na, standardized by the Sum-Peak method. The error bars in the Monte Carlo values are statistical only.

Table 1
Typical uncertainty components of activity concentration per data point (one standard deviation: $k=1$).

Components	Uncertainty (%)
Counting statistics	0.05
Weighing	0.11
Dead time	0.01
Background	0.13
Resolving time	0.05
Extrapolation of efficiency curve (Sum-Peak)	0.08
Extrapolation of efficiency curve (Nuclear-Peak)	0.16
Combined uncertainty (Sum-Peak)	0.10
Combined uncertainty (Nuclear-Peak)	0.18
Combined uncertainty, parameter h	0.19
Combined uncertainty, parameter $(1-h)/h$	2.1

The errors in the extrapolation curves include all data points.

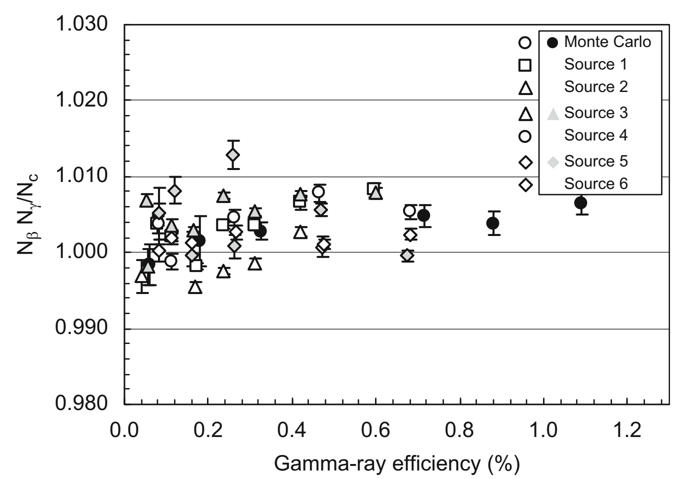

Fig. 5. Extrapolation curve for ^{22}Na, standardized by the Nuclear-Peak method. The error bars in the Monte Carlo values are statistical only.

branching ratio) is very sensitive to small variations in ***h***. Since the experimental beta efficiencies are in the range of 98.0%–99.9%, the difference in the experimental and the Monte Carlo slopes yields negligible change in the final activity value which resulted: 685.3(6) kBq mg^{-1}, using a value of 0.1044(4) as the EC/β^+ branching ratio (Moura and Nuevo, 1973).

Fig. 5 shows the extrapolation curve obtained for the Nuclear-Peak method. Several radioactive sources are plotted together with the Monte Carlo calculation. Although the fluctuations are in some cases larger than the error bars estimated by propagation, the overall behavior follows approximately the curve shape predicted by Monte Carlo. From this curve it was possible to obtain the average activity of the solution: 685.0(12) kBq mg^{-1} in reasonable agreement with the original value of 682.4(16) kBq mg^{-1} (Moura and Nuevo, 1973). With this new activity value, obtained by means of Monte Carlo calculation, the new EC/β^+ branching ratio resulted: 0.1039(21). This result was calculated from the value of ***h***, which in turn was taken from the ratio of extrapolation values from the two methods, namely: Sum-Peak (***h.N***$_0$) and Nuclear Peak (***N***$_0$). This branching ratio value is in reasonable agreement with those from recent literature, as follows: 0.1075(25) (Sy′ kora and Povinec, 1986), 0.1050(29) (Kunze et al., 1990) and 0.1084(27) (Nähle et al., 2008).

4. Conclusion

Theoretical predictions by Monte Carlo simulation of extrapolation curves obtained in the standardization of ^{22}Na by the Sum-Peak and Nuclear-Peak methods were performed. A new approach was developed in order to take into account the effect of annihilation photon emission occurring far from the radioactive source. This effect yields a variation in the gamma-ray efficiency of NaI crystals, changing the behavior of the extrapolation curve. In the present paper this effect was considered by replacing the usual gamma-ray response table at 1022 keV, with the one calculated by MCNPX using positron as a radiation source at average positron energy of 215.6 keV.

The total gamma-ray efficiency was calculated for each positron energy sorted by Monte Carlo code ESQUEMA, by applying a sigmoidal function. The theoretical results applied to ^{22}Na were in good agreement with the experimental extrapolation curves. From these results it was possible to obtain a new EC/β^+ branching ratio of 0.1039(21) in reasonable agreement with those taken from recent literature.

Using the present version of code ESQUEMA, the Monte Carlo simulation for ^{22}Na was possible because Auger electrons have energies below 1 keV and were completely absorbed by thin films placed on the radioactive source. Further refinements are foreseen to this code in order to allow application to other EC/β^+ emitters, in which Auger electrons may have higher energies and will not be absorbed by thin films. In these cases, two response tables for the beta detector should be used: one for electrons and another one for positrons.

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