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STANDARDIZATION OF Y-90 BY TRACING METHOD

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ABSTRACT

This paper describes the procedure followed by the Nuclear Metrology Laboratory (LMN) at IPEN - CNEN/SP, in São Paulo, for the standardization of the ^{90}Y, undertaken by the tracer technique using the ^{24}Na beta-gamma emitter as the tracer. The measurements were carried out, by means of a $4\pi\beta-\gamma$ coincidence system. For the observed events registration was used the TAC method. A Monte Carlo simulation for generating the extrapolation curve was applied to obtain the correction of the efficiency for determining the solution activity. The correction factor of the efficiency was also calculated by means of a semi-empirical formula. The ^{90}Y activity results obtained by both methods were compared.

1. INTRODUCTION

The standardization of ^{90}Y performed by the Nuclear Metrology Laboratory (LMN) at IPEN, in São Paulo by means of the tracing method is described [1]. The radionuclide ^{90}Y was chosen to be standardized with accuracy in a primary system due to its importance in the field of nuclear medicine, where it is used as a therapeutic agent on a monoclonal antibody, for targeting cancer cells or as the active ingredient in glass microspheres used in treating liver tumors [2]. The ^{90}Y has a half-life of 2.7 days, it decays with 99.98 % by beta emission with the end point energy of 2279.8 keV to the ground state of ^{90}Zr [3]. In Fig.1 the ^{90}Ydecay scheme is presented.

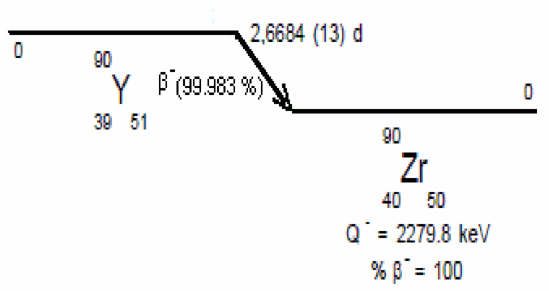

Figure 1: Simple decay scheme of ^{90}Y (all energies in keV) [3].

The measurement was performed in 4πβ–γ coincidence system, measuring the pure beta emitter mixed with a beta-gamma emitter, which provides the beta detection efficiency. ^{24}Na which decays with half life of 0.623 day [3] by beta particle, with end point energy of 1393 keV, followed by two gamma-rays, was used as tracer. The beta efficiency was obtained by selecting the 1369 keV total absorption peak at the gamma channel, the ^{24}Na decay scheme is showed in Fig. 2.

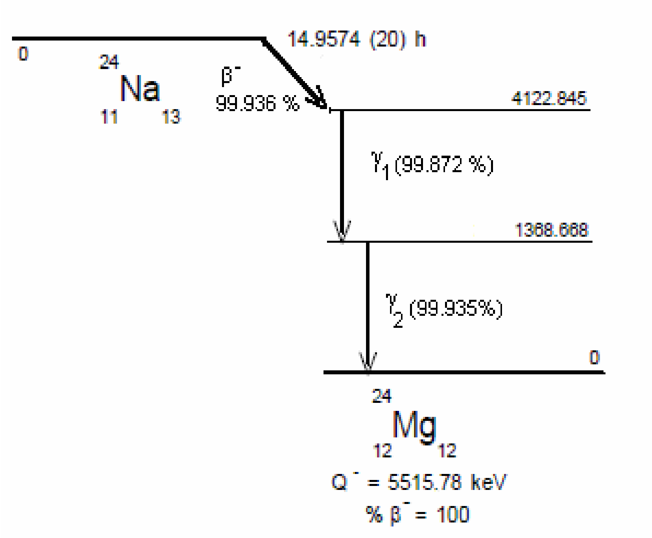

Figure 2: Simple decay scheme of ^{24}Na (all energies are in keV).

The electronic system used was the conventional for this kind of measurement [4]. The observed events were registered using a Time to Amplitude Converter (TAC) associated with a Multichannel Analyzer [5]. The activity was calculated by the Contact code, developed at LMN [6]. A Monte Carlo simulation [7,8], in which is possible to determine the extrapolation curve to obtain the correction for the efficiency was accomplished. Moreover, to confirm the solution activity determined by using the Monte Carlo prediction, a corrected factor *r* obtained by a semi-empirical formula [9] presented in 2.2 section was applied for determining the activity with Contact code.

2. EXPERIMENTAL METHOD

2.1. Source Preparation

The ^{90}Y solution was supplied by the IPEN Radiopharmaceutical Center. This solution was diluted in 0.001 g/L EuCl3 in 0.1M HCl. The ^{24}Na was obtained by the ^{23}Na(n,γ)^{24}Na reaction, by means of the irradiation of 1 mg of $NaNO_3$ in a thermal neutron flux at the IPEN research reactor IEA-R1. To prepare the ^{24}Na radioactive solution, the $NaNO_3$ irradiated was diluted in 0.1 M HNO_3. For the measurements of the beta pure, nine radioactive sources were prepared with a mix of ^{24}Na and ^{90}Y by dropping known aliquots into a Collodion film substrate 20 μg cm^{-2} thick. This film was previously coated with a 10 μg cm^{-2} gold layer in order to make the film conductive. On the sources was deposited a seeding agent (CYASTAT) to improve the uniformity of the sources, which were dried in a nitrogen jet at 45 ºC. For the ^{24}Na standardization six sources were prepared as same the procedure described.

2.2. 4πβ−γ Coincidence method

The method of coincidence is a primary method because it does not depend on the parameters of radionuclide decay scheme. The 4πβ−γ coincidence system consists of a 4π proportional counter (PC) filled with P-10 gas, operated at 0.1 MPa coupled to a NaI(Tl) scintillator. This method is applied mainly to the nuclides which disintegrate by two different radiation emission α-γ, β-γ, EC-γ and e-X ray.

In the tracing method, the 4πβ−γ coincidence system is used in the standardization of radionuclides which decay by a single type radiation. In this method, it is necessary a mixture of a pure β emitter with an emitter β-γ, whose end point energies beta are similars. The emitter β-γ acts as a tracer [1] given the beta efficiency.

When the β−γ and the β-emitter are combined in a single source, a functional relationship exists between the detection efficiencies [9], which can be defined by a polynomial function G where:

$$\left(\frac{1-\varepsilon_{\beta P}}{\varepsilon_{\beta P}}\right) = G\left(\frac{1-\varepsilon_{\beta T}}{\varepsilon_{\beta T}}\right) \tag{1}$$

The equation of the tracer method can be written similarly to the method of coincidences as:

$$\frac{N_\beta N_\gamma}{N_C} - N_{OT} = N_{OP}\left(1 + G'\left(\frac{1-\varepsilon_{\beta T}}{\varepsilon_{\beta T}}\right)\right) \tag{2}$$

Where

P and T correspond to a pure beta emitter and tracer, respectively;

N_{OP} is the rate of disintegration of a pure beta;

N_{OT} is the rate of disintegration of a tracer previously determined;

N_β is the 4π (PC) count rate of pure beta plus tracer;

N_γ is the count rate gamma of tracer;

N_c is the count rate of coincidences;

$\varepsilon_{\beta P}$ is the detection efficiency of the pure beta;

$\varepsilon_{\beta T}$ is the detection efficiency of the tracer;

G' function is adjusted by extrapolation technique that relates the beta pure efficiencies, and efficiency of the tracer.

The calibrations of the tracer and the tracer plus pure beta were performed by selecting the 1369 keV gamma-ray of ^{24}Na in the gamma detector.

In this paper the extrapolation curve $(N_\beta N_\gamma / N_c - N_{0T})$ versus $(1- N_c / N_\gamma) / N_c / N_\gamma$ used for determining the activity was those predicted by the Monte Carlo simulation.

The CONTACT code besides the ratio $N_\beta N_\gamma / N_c$ related to each efficiency parameter N_c / N_γ, it also calculates the correction factor r by applying a semi-empirical formulae [10], which establishes a relationship between the efficiency of the tracer and efficiency of the pure beta emitter considering the end-point beta-ray energy of each radionuclide.

This function is give by:

$$r = \frac{\varepsilon_{\beta T}}{\varepsilon_{\beta P}} = exp\left\{ - ln\left(\varepsilon_{\beta T}\right)\left[1 - \left(\frac{E_T}{E_P}\right)^{1.14}\right]\right\} \qquad (3)$$

Where:

$\varepsilon_{\beta T}$ is the detection efficiency of the tracer beta;

$\varepsilon_{\beta P}$ is the detection efficiency of the pure beta;

E_T is the end-point beta-ray energy of tracer;

E_P is the end-point beta-ray energy pure beta;

2.3. Monte Carlo Simulation

The counting rates for β, γ and coincidence can be calculated through Monte Carlo simulation using MCNP5 (Monte Carlo N-Particle Transport Code System) [7] that needs as input some data about the source, system geometry and detector materials in order to calculate the efficiency. These data are used by a code called ESQUEMA [8], developed in the LMN/IPEN, that allows to change the efficiency of the detectors by simulating external absorbing materials and electronic discrimination. The technique of linear extrapolation of the efficiency can be used to calculate the activity value of the given source.

3. RESULTS

Fig. 3. Shows experimental values of $(N_\beta N_\gamma / N_c - N_{0T})$ as a function of $(1-N_c / N_\gamma)/N_c / N_\gamma)$ obtained by Contact code (open circles) compared to the normalized Monte Carlo calculation simulating the absorber (close circles). The beta efficiency was varied from 98 % to 93 %.

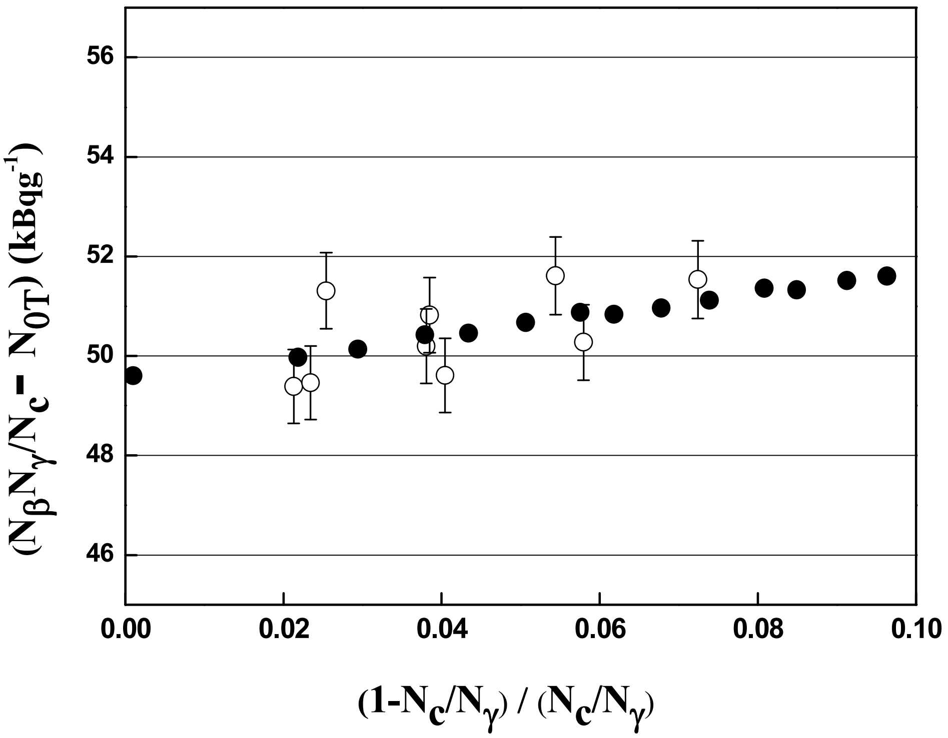

Figure 3: Extrapolation curve of ^{90}Y obtained using simulation with absorbers (close circles) and experimental data (open circles).

Table. 1 shows the experimental data $(N_\beta N_\gamma/N_c - N_{0T})$ and the efficiency parameter $(1-N_c/N_\gamma)/N_c/N_\gamma)$. Uncertainties involved are shown in parenthesis. In Table 2 the values of the activities corrected by the Monte Carlo (M.C.) extrapolation curve and by applying the ***r*** factor are presented.

Table 1: Values of ($N_\beta N_\gamma/N_c - N_{0T}$ and efficiency parameter ($1-N_c/N_\gamma)/N_c/N_\gamma$) for the nine sources measured

Source	($N_\beta N_\gamma/N_c - N_{0T}$) x 10^3 (cps g^{-1})	($1-N_c/N_\gamma)/N_c/N_\gamma$)
1	49.39 (7)	0.021 (2)
2	49.46 (7)	0.023 (3)
3	51.31 (8)	0.025 (2)
4	50.20 (7)	0.038 (2)
5	50.82 (8)	0.038 (2)
6	51.61 (8)	0.054 (3)
7	49.61 (7)	0.040 (3)
8	50.27 (7)	0.058 (3)
9	51.54 (8)	0.072 (3)

Table 2: Values of the activities corrected by the Monte Carlo (M.C.) extrapolation curve and by applying the *r* factor. .

Source	Activity by M.C. (kBq.g^{-1})	Activity by *r* factor (kBq.g^{-1})
1	49.99 (7)	50.00 (7)
2	50.85 (8)	50.76 (7)
3	49.38 (7)	49.40 (7)
4	49.03 (7)	48.94 (7)
5	50.10 (8)	50.01 (7)
6	50.41 (8)	50.50 (7)
7	49.06 (7)	48.97 (7)
8	48.79 (7)	48.77 (7)
9	49.02 (7)	49.07 (7)
Average weighted	49.60 (2)	49.58 (2)

3. CONCLUSIONS

The activity obtained by the two methods is in agreement within the experimental uncertainty, showing that both methods can be applied to standardize the ^{90}Y radionuclide by the tracer method. New measures will be made with external absorbers to vary the beta efficiency and the results will be compared with the Monte Carlo prediction.

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