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TRACING METHOD APPLIED TO THE STANDARDIZATION OF ^{137}Cs

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ABSTRACT

The procedure followed by the Laboratório de Metrologia Nuclear at the IPEN, in São Paulo, for the standardization of ^{137}Cs is described. The activity measurement was carried out in a 4πβ–γ coincidence system, by the tracing method. The radionuclide ^{134}Cs is a β–γ emitter and was chosen as tracer because of its end-point beta-ray energy which is similar to ^{137}Cs. Some sources were prepared by dropping β pure and β–γ solutions directly on Collodion film, and other sources were prepared from a ^{134}Cs + ^{137}Cs solution mixed previously in equal proportions. The activity of the solution was determined by the extrapolation technique.

1. INTRODUCTION

The Laboratório de Metrologia Nuclear (LMN) at IPEN has provided α, β and γ standard sources to several users of detection systems. To achieve this objective, the LMN has developed standardization methods for several β–γ, α–γ and X-γ, emitters, by means of 4πβ–γ coincidence system [1,2]. In the case of pure beta emitters, the LMN usually applies the tracing method. This method consists of measuring a pure beta emitter mixed with a β–γ emitter, which provides the beta detection efficiency.

Though ^{137}Cs is usually considered a β–γ emitter, the coincidence technique cannot be used because the daughter radionuclide is produced in a meta-stable state. Therefore it must be treated as a pure beta emitter for absolute calibration purposes. It decays with half-life of (30.15 ± 0.06) y [3] by beta emission with end-point energy of 1173.2 keV (5.4%) to the ground state of ^{137}Ba and by beta emission with end-point energy of 511.5 keV (94.6%) to the excited level of ^{137m}Ba This level has a meta-stable state which decays with (2.554 ± 0.002) minutes to the ground state of ^{137}Ba by 661.6keV gamma-ray emission (Fig.1).

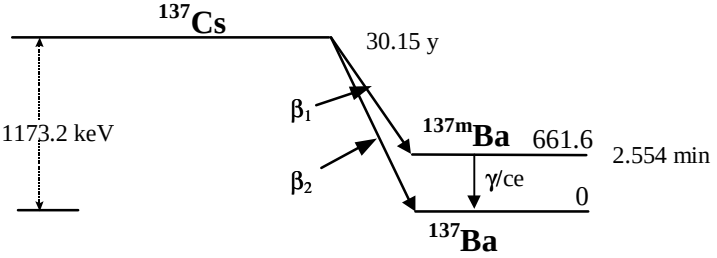

Figure 1. Decay scheme of ^{137}Cs [3].

The radionuclide ^{134}Cs was the β–γ emitter selected to be used as tracer because it has the same chemical composition and it emits beta particles with end-point energy close to those of ^{137}Cs.

2. EXPERIMENTAL METHOD

2.1. Source Preparation

The sources to be measured in the coincidence system have been prepared by dropping known aliquots of the radioactive solution on a 20 $\mu g\ cm^{-2}$ thick Collodion film. This film had been previously coated with a 10 $\mu g\ cm^{-2}$ gold layer in order to turn the film conductive. The sources were dried in dessecator. The mass determination has been performed using the pycnometer technique, in a Sartorius MC 21S balance [4]. Two kinds of sources were prepared: the first was prepared using a mixed solution of tracer and beta pure at a ratio of 1:1. The second was prepared by dropping individually known aliquots of tracer over known aliquots of pure beta solution. The sources from the mixed solution were identified as M and the other were identified as I.

2.2. $4\pi\beta$–γ Coincidence Measurement

The $4\pi\beta$–γ coincidence system used for measuring the tracer solution and the beta pure plus tracer solution, consisted of a proportional counter with 4π geometry filled with 0.1 MPa P10 gas mixture, coupled to a pair of 76mm x 76 mm NaI(Tl) crystals. The measurements were performed by integral mode for beta detection, cutting off only the electronic noise. The calibrations of tracer and tracer plus pure beta were performed selecting a γ–ray discrimination window in the (796 + 802) keV energy range. In this window the decay scheme correction is expected to be small.

The coincidence equations applied to ^{134}Cs +^{137}Cs are given by:

$$N_\beta = \left\{ N_{0tr} \left[\varepsilon_{\beta tr} + \left(1 - \varepsilon_{\beta_{tr}}\right) \frac{\alpha_{tr} \varepsilon_{ce_{tr}} + \varepsilon_{\beta\gamma_{tr}}}{1 + \alpha_{tr}} \right] \right\} + N_{0p} \left(\varepsilon_{\beta p} + C\right) \tag{1}$$

$$N_\gamma = N_0 \varepsilon_{\gamma tr} \frac{1}{1 + \alpha_{tr}} \tag{2}$$

$$N_c = N_0 \varepsilon_{\beta tr} \varepsilon_{\gamma_{tr}} \frac{1}{1 + \alpha_{tr}} \tag{3}$$

Equations (1), (2) and (3) lead to

$$\frac{N_\beta N_\gamma}{N_c} = N_{0tr}\left\{1 + \frac{\left(1-\varepsilon_{\beta tr}\right)}{\varepsilon_{\beta tr}}\frac{\left(\alpha\varepsilon_{ce_{tr}} + \varepsilon_{\beta\gamma tr}\right)}{1+\alpha}\right\} + N_{0p}\frac{\varepsilon_{\beta p}}{\varepsilon_{\beta tr}} + \frac{C}{\varepsilon_{\beta tr}} \qquad (4)$$

Where:

N_β, N_γ and N_c are beta, gamma and coincidence counting rates, respectively;

N_{0tr} is the tracer radioactive source disintegration rate;

N_{0p} is the pure beta radioactive source disintegration rate;

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

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

$\varepsilon_{\gamma_{tr}}$ is the gamma detection efficiency;

$\varepsilon_{\beta\gamma_{tr}}$ is the tracer gamma detection efficiency for beta detector;

ε_{cetr} is the tracer conversion electron detection efficiency;

α_{tr} is the tracer total internal conversion coefficient;

$C = \frac{b}{(1+\alpha)_p}\left(\alpha_p\varepsilon_{ce} + \varepsilon_{\beta\gamma_p}\right)$ is the contribution of conversion electrons and unconverted gamma-ray counted in the beta detector [5]:

Where:

b is the probability of beta emission to the metastable level;

$\varepsilon_{\beta\gamma p}$ is the gamma detection efficiency of beta detector for the pure beta;

ε_{ce} is conversion electron detection efficiency associated to the pure beta;

α_p is the total internal conversion coefficient associated to the pure beta;

These parameters were taken from the literature and are shown in Table 1.

Table 1. Factor C; values of b, α_t and $\varepsilon_{\beta\gamma}$ from the literature

Reference	Parameters	Values	C
[5]	b (branch β^- to ^{137m}Ba)	0.9461± 0.0023	0.09568 ± 0.0007
[3]	α_t (of ^{137m}Ba)	0.1097 ± 0.0010	
[9]	$\varepsilon_{\beta\gamma}$ (for γ -ray of 661.6 keV)	0.0025 ± 0.0002	

Since the tracer disintegration rate is previously known, equation (4) becomes

$$\frac{N_\beta N_\gamma}{N_c} - N_{0tr} = N_{0P}\frac{\varepsilon_{\beta P}}{\varepsilon_{\beta tr}} + \frac{C}{\varepsilon_{\beta tr}} \qquad (5)$$

When the beta pure and the tracer are combined in a single source, a relationship between the detection efficiencies can be represented by a function F of the tracer efficiency [6]. This relation can be defined by

$$\varepsilon_{\beta p} = F\left(1-\varepsilon_{\beta tr}\right) \qquad (6)$$

Or by a function G given by

$$\varepsilon_{\beta p} = G\left(\frac{1-\varepsilon_{\beta tr}}{\varepsilon_{\beta tr}}\right) \tag{7}$$

where $\varepsilon_{\beta tr}$ is approximately equal to the efficiency parameter N_c/N_γ .

Expression (5) can be rewritten as:

$$\frac{N_\beta N_\gamma}{N_c} - N_{0tr} = N_{0p}\left(1 + C + G'\left(\frac{1 - N_c/N_\gamma}{N_c/N_\gamma}\right)\right) \tag{8}$$

The function G' is usually represented as a polynomial of the inefficiency , and goes to unity when $(1-N_c/N_\gamma)/\ N_c/N_\gamma$ goes to zero. Solving equation (8) by polynomial least square fitting, the linear coefficient yields the $N_{0p}(1+C)$ value.

3. RESULTS AND DISCUSSION

The tracer specific activity was previously obtained by averaging ten radioactive sources values measured in the coincidence system. The tracer decay scheme correction factor used was (0.0097 ± 0.0019)[7].

The pure beta activity was determined by the extrapolation technique, changing the tracer efficiency parameter N_c/N_γ by using Collodion external absorbers over and under the sources. Fig. 2 shows the extrapolation curve obtained with sources prepared from solution M and Fig. 3 shows the corresponding curve obtained with sources prepared by dropping aliquots individually (I).

The solid curves were obtained by Linear Least square fitting of a second degree polynomial using code LINFIT[6], which incorporates covariance matrix methodology. Table 2 presents the intercept values, $N_0(1+C)$, obtained for the two curves. The final ^{137}Cs disintegration rate was obtained dividing the intercept by the (1+C) value shown in Table 1. The value of factor C determined from literature data is presented in Table 1.

Table 2. Intercept obtained by linear least square fitting

Group	Intercept (x 10^3)	Average (x 10^3)
M	(159.0 ± 1.1)	(159.4 ± 1.0)
I	(161.3 ± 2.6)	

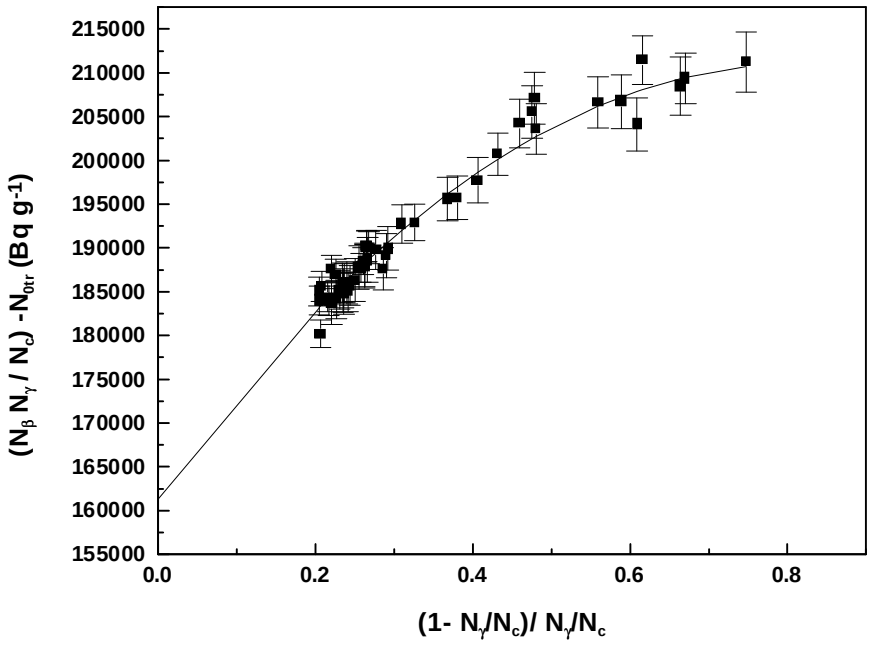

Figure 2. Extrapolation curves of $N_\beta N_\gamma/N_C$ as a function of $(1- N_C/N_\gamma)/ (N_C/N_\gamma)$ for group M sources.

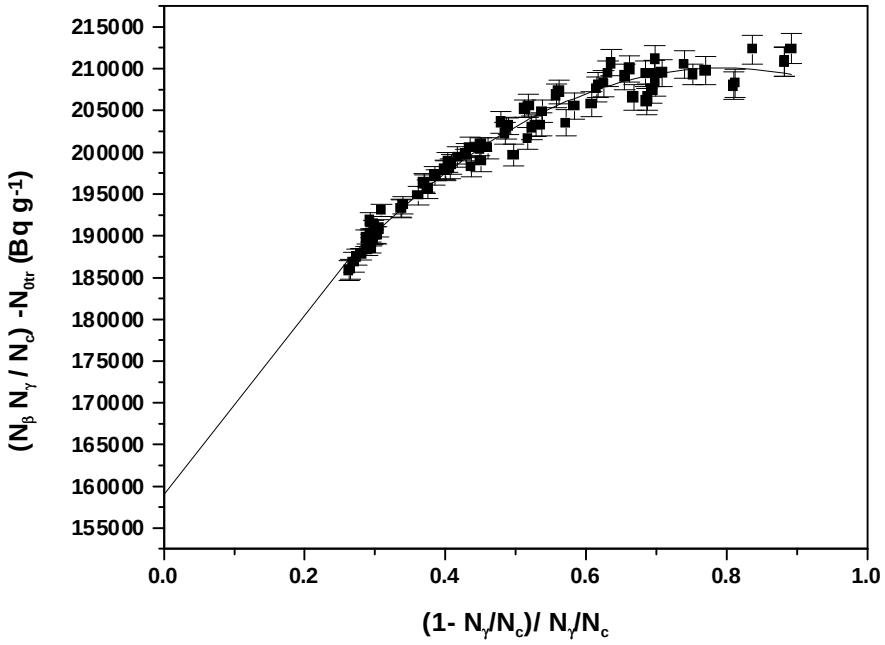

Figure 3 Extrapolation curves of $N_\beta N_\gamma/N_C$ as a function of $(1- N_C/ N_\gamma)/ (N_C/ N_\gamma)$ for group I sources.

As can be seen, both results agree within the estimated uncertainty. The final activity of ^{137}Cs solution was calculated by the unweighted average between the two preparation methods, resulting (145.6. ± 1.4) kBq g^{-1}.

4. CONCLUSION

The values of N_0 from two methods are in agreement within the experimental uncertainty. The source preparation by dropping individually from the two (pure beta and tracer) radioactive solutions on the Collodion film is much easier to perform and uses less radioactive material. For this reason it will be adopted for future measurements. A Monte Carlo simulation of the extrapolation curve is underway in order to confirm the behavior of experimental data shown in Figures 2 and 3.

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