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PRIMARY STANDARDIZATION OF ^{90}Sr-^{90}Y RADIOACTIVE SOLUTION

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ABSTRACT

In the present work, the procedure developed by the Nuclear Metrology Laboratory (LMN) at IPEN, for the primary standardization of ^{90}Sr-^{90}Y, is presented. The method applied has been the efficiency tracing technique using a 4πβ–γ coincidence system. That consists in the measurement of a beta pure emitter along with a beta-gamma emitter, previously standardized, which will provide the beta efficiency. In this work, the beta-gamma emitter used was ^{60}Co. The beta efficiency has varied using external absorbers, and the specific activity was determined using the extrapolation curve. ESQUEMA Code, which predicts the extrapolation curve by means of Monte Carlo technique, was applied, and the specific activity obtained from Monte Carlo simulation was compared with the experimental, showing good agreement within the experimental uncertainties.

1. INTRODUCTION

^{90}Sr is a nuclear fission product with half-life of 28.8 y. It decays by 100% beta minus emission, with end point energy of 546 keV to ^{90}Y, which also decays by beta minus, mainly to the ^{90}Zr ground state, with the end point energy of 2278.7 keV and half-life of 2.7 d. These radionuclides, after few weeks from the ^{90}Sr production, reach the secular equilibrium. Fig. 1 shows ^{90}Sr-^{90}Y decay scheme [1, 2].

Due to the high beta energy emitted, ^{90}Sr-^{90}Y is a radiation hazard present in fallout from nuclear weapons, nuclear accidents, in radioactive waste of nuclear plants and as an impurity in radiopharmaceuticals produced by fission reaction, such as ^{99}Mo-^{99m}Tc; then, it is important that its presence may be detected with accuracy. For this, standard solutions used to calibrate secondary systems have been standardized on primary systems.

In the present work, the procedure developed by IPEN Nuclear Metrology Laboratory (LMN), for the primary standardization of ^{90}Sr-^{90}Y, is presented. The method applied was the efficiency tracing technique, using a 4πβ–γ coincidence system [3, 4]. This method consists in the measurement of the ^{90}Sr-^{90}Y (beta pure emitters), along with a beta-gamma emitter, previously standardized, which will provide beta efficiency. The beta-gamma emitter used was ^{60}Co, which decays by beta particle, followed by two gamma rays of 1173.24 keV and 1332.51 keV, respectively. Fig. 2 shows its decay scheme [5].

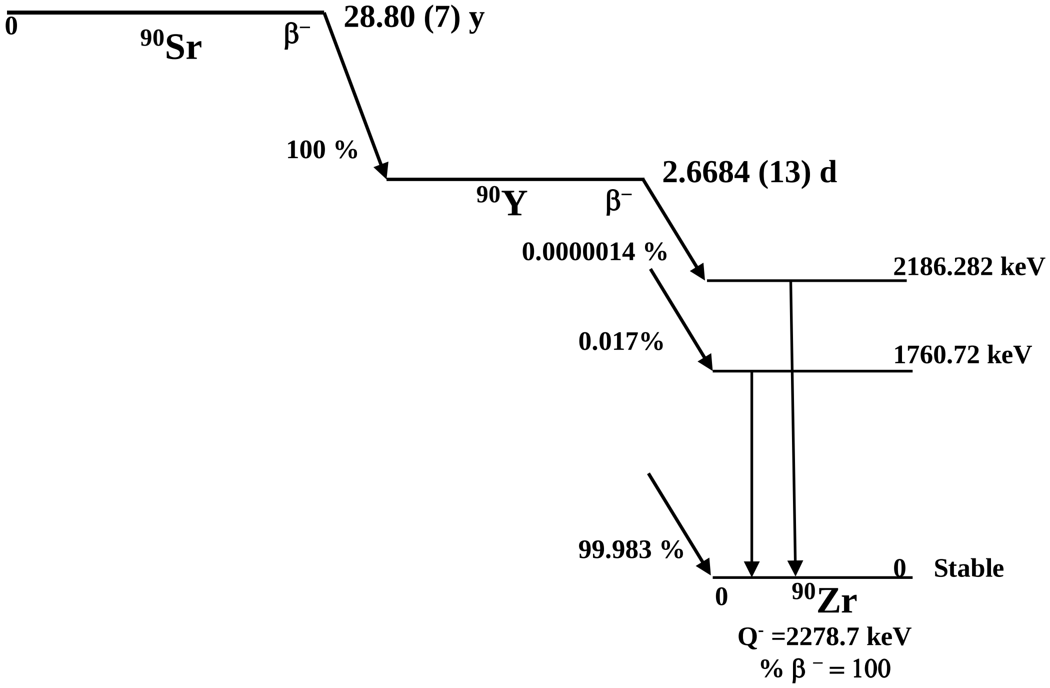

Figure 1: ^{90}Sr-^{90}Y decay scheme [1, 2]

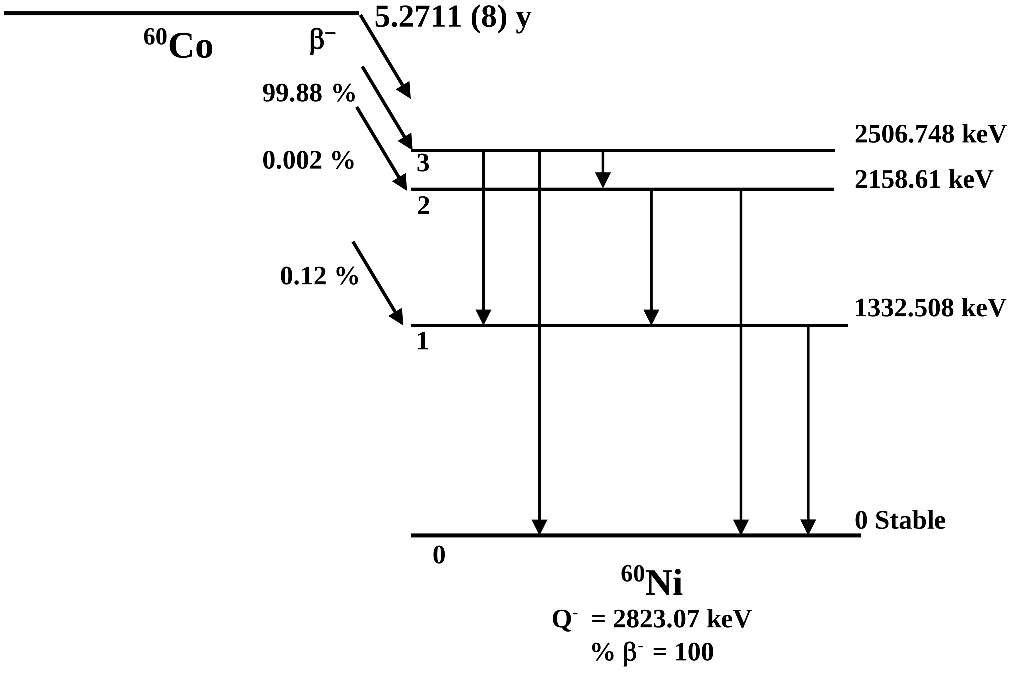

Figure 2: ^{60}Co decay scheme [4]

The measurements were carried out in the 4πβ–γ coincidence system; the events were registered using a Time to Amplitude Converter (TAC), associated with a Multi-channel Analyzer. The activity per unit mass of the solution was determined by the extrapolation technique, using external absorbers.

The experimental extrapolation curve was compared with the extrapolation curve predicted by means of ESQUEMA Code [6], which uses the Monte Carlo technique for simulating the 4πβ–γ coincidence system.

2. EXPERIMENTAL SET UP

2.1. Source preparation

For the tracing method, six sources were prepared by dropping known aliquots of each of the radioactive solutions (beta pure and beta-gamma) using, as substrate, a stainless-steel frame with a diameter of 4 cm, covered with 20 μg cm^{-2} thick Collodion film. This film was, previously, coated with a 10 μg cm^{-2} gold layer, in order to make the film conductive. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a desiccator. The sources mass was accurately determined by the pycnometer technique, in XP56 Mettler balance. ^{60}Co beta-gamma tracer was, previously, standardized by measuring six sources prepared by the same procedure. The beta efficiency varied by adding thin absorbers of Collodion films coated with gold and aluminum foils on both sides of the sources.

2.2. 4πβ–γ coincidence system

The 4πβ–γ coincidence system consists in a gas flow 4π proportional counter for beta detecting, operated at 0.1 MPa, with methane/argon gas (10 % Methane +90 % Argon), coupled to two 76 mm x 76 mm NaI(Tl) crystals, for gamma detecting. The conventional electronic system was used and the observed events were registered by TAC (Time to Amplitude Converter) method [7]. The gamma ray selected was the full absorption peak of ^{60}Co (1173.24 keV plus 1332.51 keV).

The proportional counter events detected are given by:

$$N_{\beta} = \left\{ N_{0tr} \left[\varepsilon_{\beta tr} + \left(1 - \varepsilon_{\beta_{tr}} \right) \frac{\alpha_{tr} \varepsilon_{ce_{tr}} + \varepsilon_{\beta\gamma_{tr}}}{1 + \alpha_{tr}} \right] \right\} + N_{0p1}\, \varepsilon_{\beta p1} + N_{0p2}\, \varepsilon_{\beta p2} \qquad (1)$$

$$N_{\gamma} = N_0 \varepsilon_{\gamma tr} \frac{1}{1 + \alpha_{tr}} \qquad (2)$$

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

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

$$\frac{N_\beta N_\gamma}{N_c} = N_{0tr} \left\{ 1 + \frac{\left(1-\varepsilon_{\beta tr}\right)}{\varepsilon_{\beta tr}} \frac{(\alpha \varepsilon_{ce_{tr}} + \varepsilon_{\beta\gamma tr})}{1+\alpha} \right\} + N_{0p1} \frac{\varepsilon_{\beta p1}}{\varepsilon_{\beta tr}} + N_{0p2} \frac{\varepsilon_{\beta p2}}{\varepsilon_{\beta tr}} \tag{4}$$

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_{0p1}	is the pure beta radioactive source disintegration rate of ^{90}Sr;
N_{0p2}	is the pure beta radioactive source disintegration rate of ^{90}Y;
$\varepsilon_{\beta tr}$	is the tracer beta detection efficiency;
$\varepsilon_{\beta p1}$	is the pure beta detection efficiency of ^{90}Sr;
$\varepsilon_{\beta p2}$	is the pure beta detection efficiency of ^{90}Y;
$\varepsilon_{\gamma tr}$	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.

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

$$\frac{N_\beta N_\gamma}{N_c} - N_{0tr} = N_{0P1} \frac{\varepsilon_{\beta P1}}{\varepsilon_{\beta tr}} + N_{0P2} \frac{\varepsilon_{\beta P2}}{\varepsilon_{\beta tr}} \tag{5}$$

When the beta pure and the tracer are combined in a single source, a relationship between the detection efficiencies may be represented by a function F of the tracer efficiency [3]. This relation may be defined by:

$$\varepsilon_{\beta p1,2} = F \left(1 - \varepsilon_{\beta tr}\right) \tag{6}$$

When the $\varepsilon_{\beta tr}$ tends to 1, both $\varepsilon_{\beta p1}$ and $\varepsilon_{\beta p2}$ tend to 1,

or by a function G, given by:

$$\varepsilon_{\beta p1,2} = G \left(\frac{1-\varepsilon_{\beta tr}}{\varepsilon_{\beta tr}} \right) \tag{7}$$

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

Expression (5) may be rewritten as:

$$\frac{N_{\beta}N_{\gamma}}{N_c} - N_{0tr} = (N_{0p1} + N_{0p2})\left(G'\left(\frac{1-N_c/N_{\gamma}}{N_c/N_{\gamma}}\right)\right) \quad \textbf{(8)}$$

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 squares fitting, the linear coefficient yields ($N_{0p1} + N_{0p2}$) value.

2.3. Monte Carlo Simulation

ESQUEMA Code, applied in this work, makes use of decay scheme parameters, system geometry and source characteristics. Hence, all detection processes in the coincidence system are simulated, predicting the behavior of the extrapolation curve by the Monte Carlo technique. The detector response curves were obtained by means of the MCNP6 radiation transport code (ORNL, 2006).

To apply the ESQUEMA Code for the tracing technique, the decay scheme of the tracer (^{60}Co) used in the simulation, is modified, including the decay scheme of ^{90}Sr-^{90}Y. As a result, the whole coincidence experiment can be simulated. In this way, equation (8) could be reproduced by calculation as a function of the beta efficiency, for the selected gamma-ray windows yielding the extrapolation curve for each experimental condition. In the present experiment, the beta efficiency was varied using external absorbers.

3. RESULTS

The tracer specific activity was previously obtained by measuring six radioactive sources in the $4\pi\beta-\gamma$ coincidence system. The ^{60}Co decay scheme correction factor used was (0.0038 ± 0.0038) [9]. Table 1 presents the ^{60}Co specific activity of the six sources measured and the N_c/N_{γ} parameter with uncertainties, respectively.

Table 1: ^{60}Co tracer activity with efficiencies

Source	N_c/N_{γ} %	Uncertainty %	Activity kBq g^{-1}	Uncertainty %
1	87.83	0.10	48.69	0.15
2	88.91	0.07	48.82	0.13
3	90.74	0.08	48.96	0.14
4	89.81	0.07	48.91	0.13
5	89.42	0.08	48.97	0.14
6	84.24	0.12	48.84	0.17
		Average	48.84	0.10

The pure beta activity was determined by the extrapolation technique changing the tracer efficiency parameter N_c / N_γ , by using external absorbers. The efficiency varied from 89 % to 67 %. Fig. 3 shows the extrapolation curve, the solid curve obtained by Linear Least Squares fitting using code LINFIT[10], which incorporates covariance matrix methodology. Fig. 4 presents the relative residuals in percentage between the experimental values and the fitting.

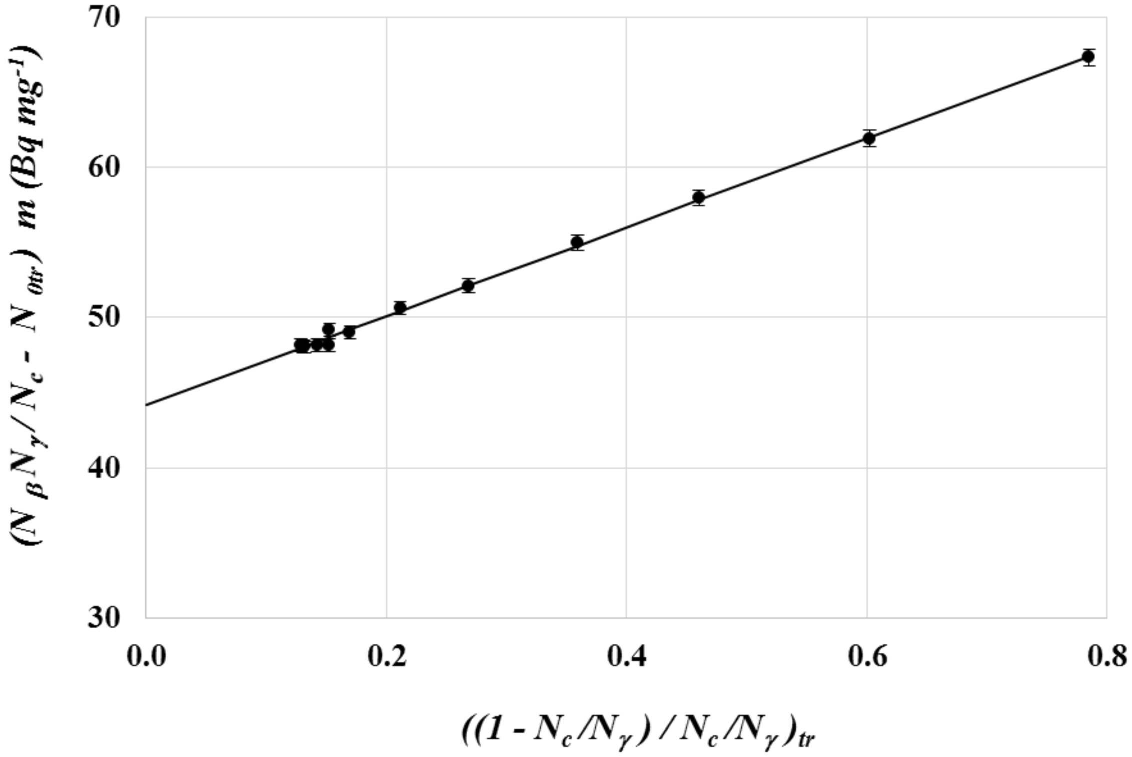

Figure 3: Extrapolation curve of ($N_\beta N_\gamma / N_C - N_{0tr}$) as a function of ($1 - N_C/N_\gamma)/(N_C/N_\gamma$). The dots with uncertainty bars are the experimental points and the continuous line, the fitted extrapolation line.

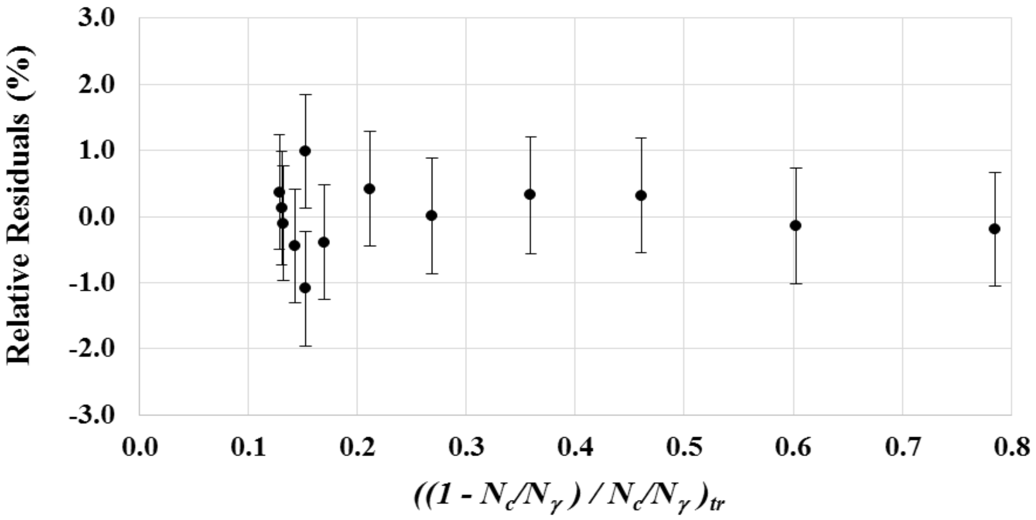

Figure 4: Relative residuals (in percent) between experimental points and the extrapolation line.

The final activity by means of the Monte Carlo simulation was calculated by Least Squares fitting combining experimental and simulated values of $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)$ for the selected gamma-ray window. The result was obtained by minimizing the following Chi-Squared value.

$$\chi^2 = (\vec{y}_{exp} - N_0\,\vec{y}_{MC})^T\, V^{-1}\, (\vec{y}_{exp} - N_0\,\vec{y}_{MC}) \qquad \textbf{(9)}$$

where:

$\vec{y}_{exp}$ is the experimental vector of $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)_{exp}$,

$\vec{y}_{MC}$ is the $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)_{MC}$ vector calculated by the Monte Carlo ESQUEMA Code, for unitary activity;

N_0 is the activity of the pure beta radioactive source;

V is the total covariance matrix, including both experimental and calculated uncertainties; and T stands for matrix transposition.

Sources activities, determined applying the correction factor obtained from the simulation curve, are presented in Table 2.

Table 2: Values of activity corrected by Monte Carlo simulation correction factor. The uncertainties correspond to one standard deviation (u=1).

Measurement	$(1-N_c/N_\gamma)/N_c/N_\gamma$	$N_\beta N_\gamma/N_c - N_{0tr}$ cps	MC Correction factor	Activity kBq g^{-1}
1	0.12862	48.16 (31)	1.0935(5)	44.04 (29)
2	0.13084	48.11 (31)	1.0951 (5)	43.93 (28)
3	0.13221	48.04 (31)	1.0961 (5)	43.83 (29)
4	0.14273	48.19 (31)	1.1035 (6)	43.66 (28)
5	0.15249	49.17 (31)	1.1105 (6)	44.28 (28)
6	0.15262	48.16 (31)	1.1106 (6)	43.74 (29)
7	0.16972	49.01 (32)	1.1231 (6)	43.64 (28)
8	0.21161	50.65 (33)	1.1522 (6)	43.96 (29)
9	0.26847	52.12 (34)	1.1918 (6)	43.73 (29)
10	0.35935	54.98 (37)	1.2546 (6)	43.83 (29)
11	0.46038	57.98 (38)	1.3229 (7)	43.83 (29)
12	0.60271	61.92 (41)	1.4177 (7)	43.68 (29)
13	0.78541	67.28 (43)	1.5381 (8)	43.75 (28)
			Average	43.84 (18)

In Table 3, the activity of ^{90}Sr-^{90}Y solution, obtained by means of the experimental fitting and the activity determined by means of MC simulation, are presented.

Table 3: Experimental activity compared with the Monte Carlo prediction

Activity (kBq g^{-1})	
Experimental	Predicted
44.18 ± 0.41	43.84 ± 0.18

These results show that activity obtained by means of Monte Carlo simulation agrees, with the experimental fit within the experimental uncertainty. This indicates that, in cases where the shape and the end-point of beta spectra of the pure beta emitters are not as similar as they have been, the Monte Carlo prediction may be used with good accuracy. Fig. 5 shows the extrapolation curve (dashed line) predicted by ESQUEMA code, normalized to the average activity calculated by Monte Carlo, compared with experimental points.

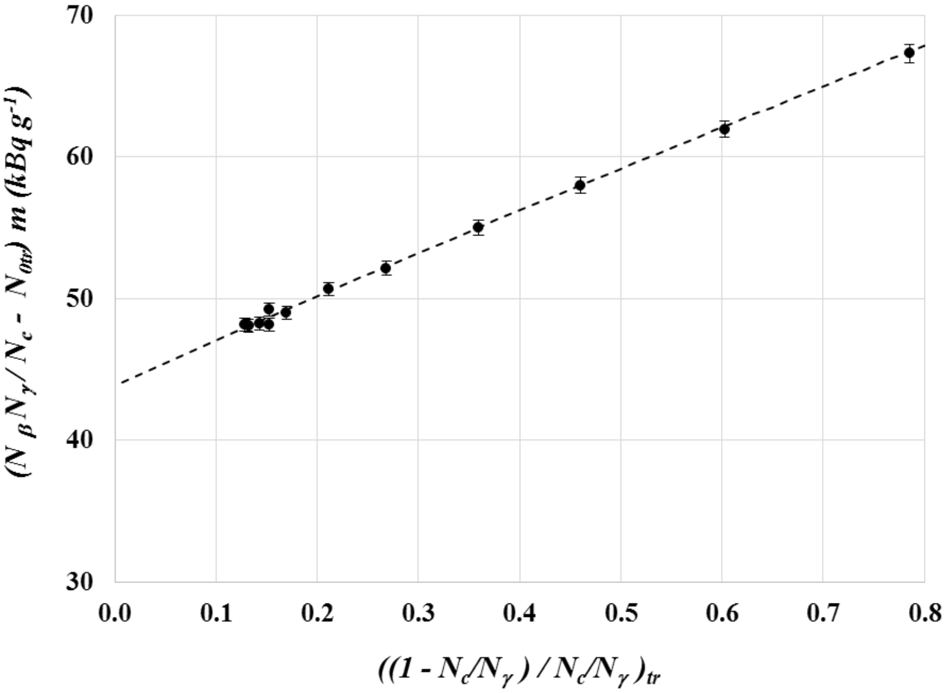

Figure 5: Extrapolation curve of ($N_\beta N_\gamma /N_C – N_{0tr}$) as a function of ($1$- N_C/N_γ)/ (N_C/N_γ). The dots with uncertainty bars are the experimental points and the dashed line represents the Monte Carlo simulation normalized to the Monte Carlo activity.

The main uncertainties involved in the tracer method using the 4πβ–γ system were: fitting procedure, counting statistics from N_β, counting beta efficiency and gamma background, which were considered uncorrelated, and the uncertainties in weighing, tracer activity, dead time, decay correction and resolving time were considered correlated. Typical partial uncertainties in percent are shown Table 4.

Table 4: Typical partial uncertainties in percent, (u=1), involved in the activity determination, with corresponding correlation.

Components	Tracer Method	Correlation
Counting statistics (included in the fitting)	-	0
Weighing ^{90}Sr-^{90}Y	0.20	1
Weighing Tracer ^{60}Co	0.20	1
Tracer activity	0.22	1
Gamma background	0.10	0
Tracer decay correction	0.05	1
Resolving time	0.10	1
Dead time	0.10	1
Extrapolation of efficiency extrapolation curve/ least squares fit error	0.60	
Shape of beta spectrum	0.60	
Combined uncertainty	0.94	

4. CONCLUSIONS

The standardization of ^{90}Sr-^{90}Y solution by means of the tracing technique in the $4\pi\beta-\gamma$ coincidence system, using ^{60}Co as tracer, succeeded and the results obtained by means of the extrapolation technique, using external absorbers and compared with the Monte Carlo simulation, have presented good agreement within the experimental uncertainty.

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