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# Primary standardization of $^{57}$Co

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ABSTRACT

This work describes the method developed by the Nuclear Metrology Laboratory in IPEN, São Paulo, Brazil, for the standardization of a $^{57}$Co radioactive solution. Cobalt-57 is a radionuclide used for calibrating gamma-ray and X-ray spectrometers, as well as a gamma reference source for dose calibrators used in nuclear medicine services. Two 4πβ–γ coincidence systems were used to perform the standardization, the first used a 4π(PC) counter coupled to a pair of 76 mm × 76 mm NaI(Tl) scintillators for detecting gamma-rays, the other one used a HPGe spectrometer for gamma detection. The measurements were performed by selecting a gamma-ray window comprising the (122 keV+136 keV) total absorption energy peaks in the NaI(Tl) and selecting the total absorption peak of 122 keV in the germanium detector. The electronic system used the TAC method developed at LMN for registering the observed events. The methodology recently developed by the LMN for simulating all detection processes in a 4πβ–γ coincidence system, by means of the Monte Carlo technique, was applied and the behavior of extrapolation curve compared to experimental data. The final activity obtained by the Monte Carlo calculation agrees with the experimental results within the experimental uncertainty.



## 1. Introduction

The radionuclide $^{57}$Co decays with a half-life of 271.80 days (Bé et al., 2004) by electron capture to the excited states of $^{57}$Fe (0.18% to 706.42 keV level and 99.82% to 136.47 keV level), emitting three main gamma-rays with energies of 14, 122 and 136 keV. These features makes $^{57}$Co a suitable radionuclide for calibrating X-ray and gamma-ray spectrometers, as well as a gamma reference source for dose calibrators used in nuclear medicine services.

This work describes the method developed by the Nuclear Metrology Laboratory (LMN) in IPEN, São Paulo, Brazil, for the standardization of a $^{57}$Co radioactive solution. The calibration was performed by means of two 4πβ–γ coincidence systems, each one using a proportional counter (PC) as the 4πβ detector.

In the first system, the gamma-rays were detected by a pair of 7.6 cm × 7.6 cm NaI(Tl) crystals placed in a sandwich configuration with respect to the 4π detector. The other used a 20% relative efficiency HPGe as the gamma-ray detector with the end cap positioned close to the PC wall. The first system was set with a gamma window comprising both the total absorption peaks of 122 and 136 keV and the second one was set to include only the 122 keV total absorption peak.

The methodology, recently developed by the LMN (Takeda et al., 2005) for simulating all detection processes in a 4π(β,X)–γ coincidence system by means of the Monte Carlo technique, was applied to these measurements and the behavior of the extrapolation curves were compared to experimental data.

## 2. Methodology

### 2.1. Coincidence equations

Although it looks simple, the decay scheme of $^{57}$Co exhibits some difficulties when applying the efficiency extrapolation technique: the 14 keV highly converted transition ($\alpha_T$=8.5) yields low energy electrons which may have detection efficiency reduction as more absorbers are placed over and under the radioactive sources.

In order to circumvent this problem several approaches for $^{57}$Co coincidence equations were developed in the literature. Williams (1966) presents a measurement using a 4π(PC)β–γ coincidence system. The relationship between the disintegration rate and the rate of observed $\beta$, $\gamma$ and $\beta\gamma$ coincidences are given by the following equation (Throughton, 1966):

$$\frac{N_\beta N_\gamma}{N_C} = N_0\left[1+\frac{1-\varepsilon}{\varepsilon}A_T+C\frac{\varepsilon-\varepsilon_x}{\varepsilon}\right] \quad (1)$$

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where

$$\frac{N_\gamma}{N_C}=\varepsilon;\quad A_T=\sum_{r=1-3}\frac{b_r\alpha_r}{1+\alpha_r}\varepsilon_{\alpha r};\quad C=\left\{b_1\left(\frac{\varepsilon_{\gamma 1}}{\varepsilon_{\gamma 2}}-1\right)\times\frac{(1-\varepsilon_{\beta\gamma 1})}{1+\alpha_1}-\frac{b_3(1-\varepsilon_{\beta\gamma 3})}{1+\alpha_3}\right\}$$

$\alpha_r$ is the total internal conversion coefficient of the $r$-th γ-ray; $\varepsilon_{\alpha r}$ is the efficiency of the β-counter to internal conversion events from the $r$-th γ-ray; $\varepsilon_x$ is the efficiency of the β-counter to electron capture events; $\varepsilon_{\beta\gamma r}$ is the efficiency of the β-counter to the $r$-th γ-ray and $\varepsilon_{\gamma r}$ is the efficiency of the γ-counter to the $r$-th γ-ray. The efficiency ε was varied by altering the composition of the counting gas from 90% A+10% $CH_4$ to 100% $CH_4$ and by using thin gold coated VYNS films. In this measurement the main correction term is $(1-\varepsilon/\varepsilon)A_T$.

Garfinkel and Hutchinson (1966) describe a 4π(PC)β–γ experiment where a proportional counter is used for detecting electrons and X-rays and a NaI(Tl) crystal for the gamma-rays. In this case, the total coincidence rate is given by

$$\frac{N_X N_\gamma}{N_C}=N_0\left[\frac{[\varepsilon_{ec}^X+(1-\varepsilon_{ec}^X)\Gamma_{a+b}^X]\Gamma_{a+b}^\gamma}{\Gamma_{a+b}^\gamma[\varepsilon_{ec}^X+(1-\varepsilon_{ec}^X)}\;\frac{b\frac{(\alpha_{14}\varepsilon_{14ce}^X+\varepsilon_{14}^X)}{1+\alpha_{14}}}{b+\left[\frac{a\varepsilon_{136}^\gamma}{\varepsilon_{122}^\gamma}\frac{1+\alpha_{122}}{1+\alpha_{136}}\right]}\right] \tag{2}$$

where $N_0$ is the disintegration rate; $N_X$, $N_\gamma$ and $N_c$ are the events detected in the proportional counter, gamma-ray and coincidence counting rates, respectively; $a$ and $b$ are the branching ratios of the gamma energy 136 and 122 keV, respectively; $\varepsilon_{ec}^X$ is the proportional counter efficiency for electron capture events; $\varepsilon_{14\ ce}$, $\varepsilon_{122\ ce}$, $\varepsilon_{136\ ce}$ are the proportional counter efficiency for the conversion electron corresponding to the 14, 122 and 136 keV transitions, respectively; $\varepsilon_{122}^\gamma$, $\varepsilon_{136}^\gamma$ are the gamma-ray detection efficiency for the 122 and 136 keV lines, respectively; $\alpha_{14}$, $\alpha_{122}$, $\alpha_{136}$ are the internal conversion coefficient corresponding to 14, 122 and 136 keV transitions, respectively; $\varepsilon_{14}^X$, $\varepsilon_{122}^X$, $\varepsilon_{136}^X$ are the proportional counter efficiency for 14, 122 and 136 keV gamma-rays, respectively,

$$\Gamma_{a+b}^X=\left\{a\frac{(\alpha_{136}\varepsilon_{136ce}^X+\varepsilon_{136}^X)}{1+\alpha_{136}}+b\left[\frac{(\alpha_{122}\varepsilon_{122ce}^X+\varepsilon_{122}^X)}{1+\alpha_{122}}+\frac{(\alpha_{14}\varepsilon_{14ce}^X+\varepsilon_{14}^X)}{1+\alpha_{14}}-\left(\frac{(\alpha_{122}\varepsilon_{122ce}^X+\varepsilon_{122}^X)}{1+\alpha_{122}}\right)\left(\frac{(\alpha_{14}\varepsilon_{14ce}^X+\varepsilon_{14}^X)}{1+\alpha_{14}}\right)\right]\right\} \tag{3}$$

and

$$\Gamma_{a+b}^\gamma=a\frac{\varepsilon_{136}^\gamma}{1+\alpha_{136}}+b\frac{\varepsilon_{122}^\gamma}{1+\alpha_{122}} \tag{4}$$

By this method, two regions on the spectra of 122 and 136 keV gamma-ray transitions obtained with NaI (Tl) were measured in coincidence.

Smith and Stuart (1975) describe a two dimensional extrapolation method. In this approach a Ge(Li) detector is coupled to a 4π(PC) detector. The counting rates could be measured separately for 122 and 136 keV transitions due to high resolution of the gamma detector. The coincidence rate is given by

$$\frac{N_\beta}{\varepsilon_l}=N_0\left[1+A\frac{(1-\varepsilon_l)}{\varepsilon_l}+B\left(1-\frac{\varepsilon_u}{\varepsilon_l}\right)\right] \tag{5}$$

where $A=\sum_i a_i F_i$; $B=A-1+\sum_i a_i b_i(1-G_i)$; $\varepsilon_l=N_{cl}/N_{\gamma l}=\varepsilon_C+\overline{1-\varepsilon_C}\varepsilon_Y$ for the 122 keV gamma window; $\varepsilon_u=N_{cu}/N_{\gamma u}=\varepsilon_C$ for the 136 keV gamma window; $\varepsilon_Y$ is the 14 keV partially internally converted efficiency for events detected by the β counter; $a_i$ are the electron capture branching ratios; $F_i$ is the probability of any transition (except the 14 keV transition itself) from the i-th electron capture branch causing a count in the β detector; $F_i=b_iG_i+(1-b_i)H_i$ with $b_i$ the fraction of the $i$-th electron capture branch which passes through the 14 keV level; $G_i$ is the contribution from the fraction $b_i$ and $H_i$ a contribution from the remainder component. In these equations, the simultaneous extrapolation for $(1-\varepsilon_l)/\varepsilon_l$ and $(1-\varepsilon_u/\varepsilon_l)$ to zero yields the activity.

A recent standardization has been performed by Havelka and Sochorová (2008). Two coincidence systems were used: one using a PC counter coupled to an HPGe gamma-ray spectrometer, and another one using a PC counter coupled to a pair of NaI(Tl) crystals. The coincidence rate was given by

$$\frac{N_{PC}N_\gamma}{N_C}\approx N_0\left(1+\left(\frac{1-\varepsilon_{PC}}{\varepsilon_{PC}}\right)k\right) \tag{6}$$

where

$$k=\left\{\frac{b}{1+\alpha_{T122}}\left[\alpha_{T122}+\varepsilon_{PC\gamma 122}\left(1-\frac{\varepsilon'_{\gamma 122}}{\varepsilon_{\gamma 122}}\right)\right]+\frac{a}{1+\alpha_{T136}}\left[\alpha_{T136}+\varepsilon_{PC\gamma 136}\left(1-\frac{\varepsilon'_{\gamma 136}}{\varepsilon_{\gamma 136}}\right)\right]\right\} \tag{7}$$

This equation is valid if $\varepsilon_{\gamma 122}=\varepsilon_{\gamma 136}$, $\varepsilon'_{\gamma 122}\approx\varepsilon'_{\gamma 136}$, $\varepsilon_{PC\gamma 122}\approx\varepsilon_{PC\gamma 136}$. In this method, the gamma-ray window was set between 30 and 300 keV.

In the methods presented above, in general, the gamma-ray window is set comprising the 122 keV+136 keV, and the proportional counter efficiency is taken from the ratio $N_X/N_\gamma=\varepsilon$, which includes all detected events. Assuming that coefficient $C$ in Eq. (1) is zero (Williams, 1966), the disintegration rate can be obtained by extrapolating the inefficiency parameter to zero. The paper by Smith and Stuart (Smith and Stuart, 1975) shows non-linear results when using a 122 keV gamma-window and predicts an error lower than 0.1% for a wide (122 keV+136 keV) gamma-gate.

In the present work the behavior of the coincidence rate as a function of the beta efficiency is based on a Monte Carlo simulation, described in Section 2.3. The merit of this approach is to try to reproduce by simulation all processes present in the decay scheme and the interactions inside all detection system components. In this way the extrapolation curve can be calculated and applied to the experimental points. A least square fitting is performed in order to provide the extrapolated activity value $\boldsymbol{N_0}$ combining experimental and simulated data.

### 2.2. Experimental setup

Two 4πβ–γ coincidence systems were used to perform the standardization: system A consisting of a 4π proportional counter coupled to two 76 × 76 mm NaI(Tl) crystal scintillators. The measurements were performed selecting a gamma-ray window comprising the (122 keV+136 keV) total absorption energy peaks in the NaI(Tl); system B, consisting of a 4π proportional counter coupled to a HPGe detector. The measurements were performed by selecting a gamma-ray window comprising the 122 keV total absorption energy peak.

The 4π proportional counters were filled with P-10 gas (90% argon+10% methane) operated at 0.1 MPa. In system A, the efficiency was changed by electronic discrimination and by using external absorbers placed on both sides of the sources. The absorbers were made of Collodion films, previously coated with a 20 μg cm$^{-2}$ gold layer. For measurements in system B, the proportional counter efficiency was changed by using external absorbers. The electronic system is described elsewhere (Fonseca et al., 2008). The events were registered by a method developed by the LMN, using a time-to-amplitude converter (TAC), associated with a multichannel analyzer (Baccarelli et al., 2008).

The radioactive sources were prepared by dropping known aliquots of the radioactive solution onto a collodion film substrate 20 μg cm$^{-2}$ thick. This film had been previously coated with a 10 μg cm$^{-2}$ gold layer in order to make the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity; the sources were dried by a nitrogen jet at 45 °C (Wyllie et al., 1970). The mass determination was performed using the pycnometer technique (Campion, 1975).

### 2.3. Monte Carlo simulation

The ESQUEMA code (Takeda et al., 2005), developed for simulating all detection processes in a 4πβ–γ coincidence system by means of the Monte Carlo technique, was used. All the information contained in the decay scheme is included as input data. The decay processes are followed, from the precursor nucleus to the ground state of daughter radionuclide, and the deposited energy for all radiations in both detectors are determined. In this way, the code reproduces the whole coincidence experiment.

This code was used in two different conditions: (a) considering the gamma-ray detection without resolution effects and (b) including the NaI experimental energy resolution in order to obtain a gamma spectrum similar to the experimental one.

In the first condition (a) it was possible to select three gamma-ray windows: 122 keV peak only, 136 keV peak only, or both. In the second condition, both gamma-rays were selected inside the same window due to NaI broad resolution.

Fig. 1 shows the results for the first condition. It is possible to verify different behaviors of the extrapolation curves. The upper curve with higher slope corresponds to the 136 keV gamma-ray energy peak alone. For the other windows the behavior of the extrapolation curve similar but with smaller slopes. Based on these results, it was decided to perform the measurement with HPGe detector selecting the 122 keV gamma-ray peak only.

The response, for the second condition (b), is presented in Fig. 2, setting artificially the 14 keV internal conversion coefficient equal to zero. As it can be seen, when this condition is imposed, the behavior of the extrapolation curve is linear.

The upper curve in this figure, calculated with a non-zero 14 keV internal conversion coefficient, indicates that a systematic bias is obtained when a straight line is used for extrapolation. The non-linearity in the high efficiency region can be attributed to variation in the 14 keV conversion electron efficiency of the proportional counter, and the Monte Carlo model gives a more realistic behavior, avoiding this systematic error.

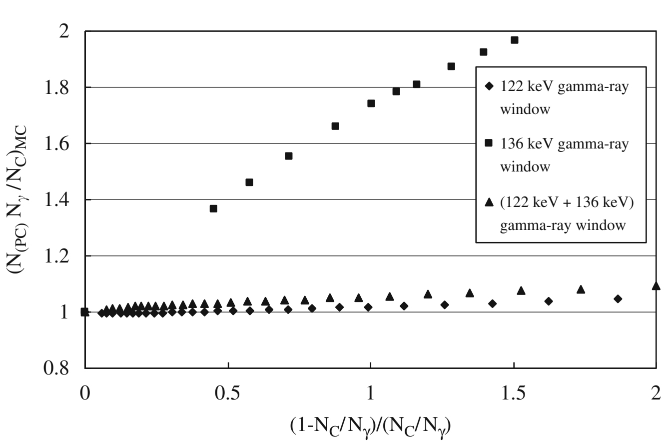


**Fig. 1.** Behavior of the extrapolation curves of $(N_\beta N_\gamma/N_C)_{MC}$ as a function of $(1-N_C/N_\gamma)/(N_C/N_\gamma)$ obtained by the Monte Carlo calculation, without energy resolution effects in the gamma detector, for the three windows selected.

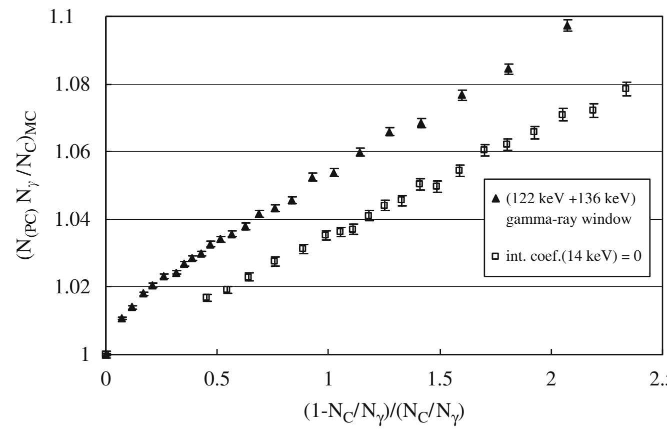


**Fig. 2.** Behavior of the extrapolation curves of $(N_\beta N_\gamma/N_C)_{MC}$ as a function of $(1-N_C/N_\gamma)/(N_C/N_\gamma)$ obtained by the Monte Carlo calculation, with energy resolution effects in the NaI gamma detector, compared with the behavior of the extrapolation curve when the decay scheme is artificially considered with the 14 keV internal conversion coefficient equal to zero.

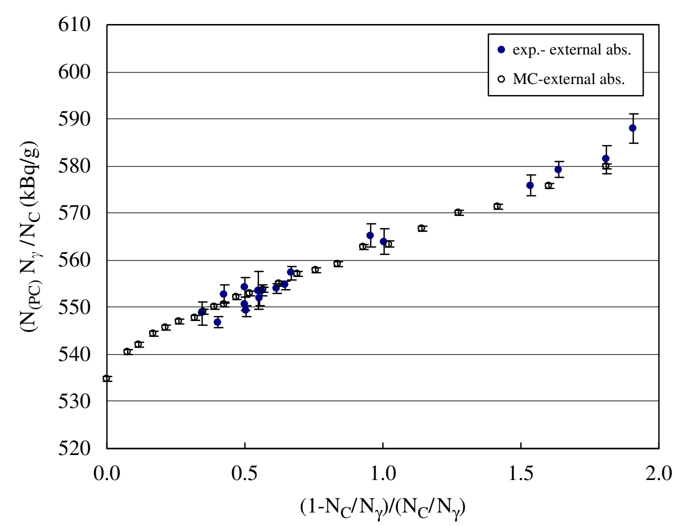


**Fig. 3.** Behavior of $N_{PC}$ $N_\gamma/N_C$ as a function of efficiency parameter $(1+N_C/N_\gamma)/(N_C/N_\gamma)$ using external absorbers. The gamma window was set including both 122 and 136 keV peaks (system A).

To compare the Monte Carlo simulation to the experimental data, a least square fit between the experimental data and the Monte Carlo calculation has been performed, according to the following equation:

$$\chi^2 = (\vec{y}_{exp} - N_0 \vec{y}_{MC})^T V^{-1} (\vec{y}_{exp} - N_0 \vec{y}_{MC}) \quad (8)$$

where $\vec{y}_{exp}$ is the experimental vector of $N_{(PC)}N_\gamma/N_C$; $\vec{y}_{MC}$ the $N_{(PC)}$ $N_\gamma/N_C$ vector calculated by Monte Carlo for unitary activity; $N_0$ the specific activity of the radioactive solution; $V$ the total covariance matrix, including both experimental and calculated uncertainties, and $T$ stands for matrix transposition.

With this code was possible to simulate variation in 4π(PC) efficiency, by changing the cut-off of deposited energy in the 4π(PC) detector and by applying absorbers on the radioactive source substrate.

## 3. Results

Figs. 3 and 4 show the measurements obtained with system A, using external absorbers and electronic discrimination,

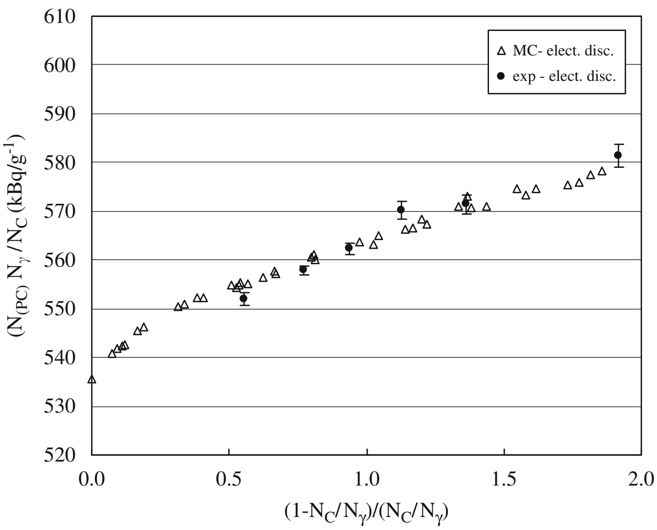


**Fig. 4.** Behavior of $N_{PC}$ $N_\gamma/N_C$ as a function of efficiency parameter $(1+N_C/N_\gamma)/(N_C/N_\gamma)$ changing the efficiency by electronic discrimination compared to the Monte Carlo calculation. The gamma window was set including both 122 and 136 keV peaks (system A).

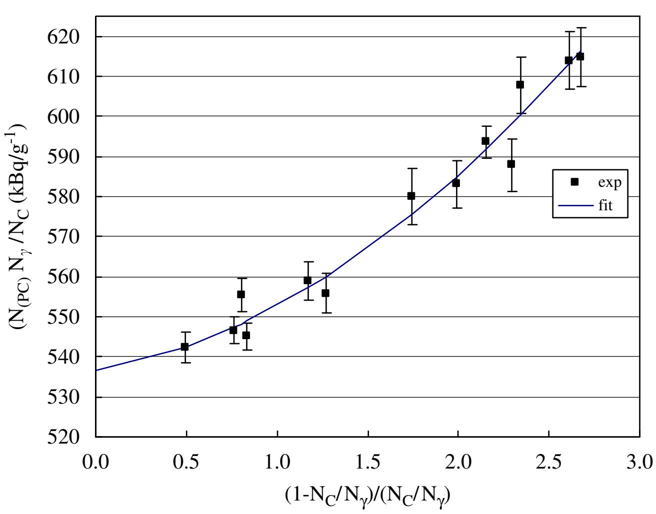


**Fig. 5.** Extrapolation curve of $N_{(PC)}$ $N_\gamma/N_C$ as a function of efficiency parameter $(1-N_C/N_\gamma)/(N_C/N_\gamma)$ using external absorbers and 122 keV gamma-ray window (system B).

respectively, in order to change the proportional counter efficiency are compared with Monte Carlo calculation. The experimental data obtained with system B, using external absorbers for changing the proportional counter efficiency, are presented in Fig. 5.

The activity concentration, using systems A and B, was obtained by means of a second degree polynomial fit to experimental data, using the code LINFIT (Dias, 1999), which incorporates the covariance matrix methodology.

Table 1 presents the extrapolated activity concentration results obtained with system A in comparison with Monte Carlo calculation. These values are in agreement within the experimental uncertainty. In this table, the extrapolated activity concentration obtained with system B is also given.

The main uncertainties involved in the measurement were counting statistics, from $N_\beta$ and $N_c/N_\gamma$ counting, weighing, dead time, decay correction, resolving time, and extrapolation curve efficiency. All these uncertainties were included in the fitting, using the covariance matrix methodology. An uncertainty budget

**Table 1**
Results of activity concentration obtained using systems A and B.

| System | Window | Activity concentration (kBq g$^{-1}$) | |
|---|---|---|---|
| | 122 keV+136 keV | Experimental | Monte Carlo |
| A | External absorber | (536.1 ± 2.2) | (534.7 ± 1.0) |
| A | Electronic discrimination 122 keV | (535.6 ± 2.9) | (535.0 ± 1.7) |
| B | External absorber | (536.5 ± 7.4) | |

**Table 2**
Uncertainty budget associated with the experimental activity concentration.

| Component | Uncertainty (%) | Comments |
|---|---|---|
| Counting statistics | 0.10 | Statistics in $N_\beta$ and $N_C/N_\gamma$ |
| Weighing | 0.10 | Balance certificate |
| Dead time | 0.05 | Pulser method |
| Background | 0.10 | Statistics counting |
| Decay | 0.01 | Half-life |
| Gandy effect | < 0.05 | Because the real coincidence region was selected by the TAC module |
| Resolving time | 0.10 | Statistics in accidental coincidence corrections |
| Extrapolation of efficiency curve | 0.35 | Least square fit error |
| Combined uncertainty | 0.41 | |

associated with the experimental activity concentration is presented in Table 2.

## 4. Conclusion

The extrapolation curve obtained with Monte Carlo simulation agrees with 4π(PC)–NaI(Tl) experimental data and confirms the conclusions made by Smith and Stuart (1975). Fig. 2 shows that for measurements at high beta efficiencies (close to 100%) the extrapolation can be performed by a straight line. However, when the efficiency is far from 100%, the fitting line may result in values slightly larger, and the extrapolation becomes more difficult. In this case the Monte Carlo prediction can be used with good accuracy.

The activity result obtained with the 4π(PC)–HPGe system is also in agreement within the experimental uncertainty. The Monte Carlo simulation for this system is underway. These results will be compared with those obtained by means of a software coincidence counting system, which is in progress. This method will provide the application of bi-parametric extrapolation, and select multiple gamma windows.

## References

Baccarelli, A.M., Dias, M.S., Koskinas, M.F., 2008. Standardization of F-18 by means of 4π(PS)-gamma plastic scintillator coincidence system. IEEE Trans. Nucl. Sci. 55 (3), 1767–1773.
Bé, M.-M., et al., 2004. Tables de Radionucléides, Monographie BIPM-5, ISBN 92-822-2204-7.
Campion, P.J., 1975. Procedures for accurately diluting and dispensing radioactive solutions. Bureau International des Poids et Mesures, Monographie BIPM-1.
Dias, M.S., 1999. LINFIT: a code for linear least square fit with covariance analysis. Internal Report, IPEN-CNEN/SP.
Fonseca, K.A., Koskinas, M.F., Dias, M.S., 2008. Measurement of gamma-ray probability per decay of Re-186. In: Proceedings of Nuclear Science Symposium, IEEE Conference, Dresden, CD-ROM.
Garfinkel, S.B., Hutchinson, J.M.R., 1966. The standardization of Cobalt-57. Int. J. Appl. Radiat. Isot. 17, 587–593.

Havelka, M., Sochorová, J., 2008. Standardization of $^{56}$Co and $^{57}$Co using software coincidence counting system. Appl. Radiat. Isot. 66, 914–918.

Smith, D., Stuart, L.E.H., 1975. An extension of the 4πβ–γ coincidence technique: two dimensional extrapolation. Metrologia 11, 67–72.

Takeda, M.N., Dias, M.S., Koskinas, M.F., 2005. Application of Monte Carlo simulation to Cs-134 standardization by means of 4π beta-gamma coincidence system. IEEE Trans. Nucl. Sci. 52 (5), 1716–1720.

Throughton, M.E.C., 1966. The absolute standardization of Cobalt-57. Int. J. Appl. Radiat. Isot. 17, 145–150.

Williams, A., 1966. Measurements of the decay scheme correction in the absolute standardization of Cobalt-57. Int. J. Appl. Radiat. Isot. 17, 366–367.

Wyllie, H.A., Johnson, E.P., Lowenthal, G.C., 1970. A procedure for stirring aliquots of radioactive solutions when making thin 4π counting sources. Int. J. Appl. Radiat. Isot. 21, 497.