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Standardization of ^{99m}Tc by means of a software coincidence system

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ABSTRACT

The procedure followed by the Nuclear Metrology Laboratory, at IPEN, for the primary standardization of ^{99m}Tc is described. The primary standardization has been accomplished by the coincidence method. The beta channel efficiency was varied by electronic discrimination using a software coincidence counting system. Two windows were selected for the gamma channel: one at 140 keV gamma-ray and the other at 20 keV X-ray total absorption peaks. The experimental extrapolation curves were compared with Monte Carlo simulations by means of code ESQUEMA.

1. Introduction

The Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear—LMN) at the IPEN, in São Paulo, has two $4\pi\beta$–γ coincidence systems composed of a gas-flow and a pressurized 4π proportional counter, respectively. The latter can also be operated as a gas-flow counter and a thin window may be provided for extracting X-rays or low energy gamma-rays to be measured by a gamma channel composed of one or a pair of NaI(Tl) scintillation counters. These scintillators can be replaced by a HPGe detector, when necessary.

One of the research areas being developed by LMN is the primary standardization of radionuclides applied to Nuclear Medicine. In this context, ^{99m}Tc became very important due to its application as a radioactive tracer and because it is commercially produced by IPEN. For this reason, it became necessary to develop a primary method for the determination of its disintegration rate.

The radionuclide ^{99m}Tc decays with a half-life of 6.0067(10) h, mainly by isomeric transition to ^{99}Tc, as shown in Fig. 1 (Bé et al., 2004). Weak beta minus transitions ($<0.003\%$) have also been observed but they were neglected in the present experiment. The most intense gamma-transitions are 2.1726 keV (99.0%) and 140.511 keV (99.0%). The first one is highly converted and the α_T coefficient is equal to $135(4)\times 10^8$ (Bé et al., 2004). For the latter gamma-transition α_T is 0.119(3). Several internal conversion transitions yields X-rays in the 2–3 keV and 18–21 keV ranges, as well as Auger electrons in the 1.6–2.9 keV and 14–21 keV ranges. Conversion electrons are emitted in 1.6–2.1 keV and 119–140 keV energy ranges.

The absence of beta emission and electron capture processes prevents the use of conventional $4\pi\beta-\gamma$ coincidence measurements for the standardization of this radionuclide. Therefore, in the present paper, the coincidence events were selected by two different methods (Goodier and Williams, 1966; Ayres and Hirshfeld, 1982; Sahagia, 2006): between 119 and 140 keV conversion electrons and 18–21 keV X-rays, and between 1.6 and 2.1 keV conversion electrons and 140.511 keV gamma-rays.

In the first method, only the disintegration rate associated with the 140.511 keV internal conversion process is measured, and the result must be divided by the conversion emission probability per decay, in order to obtain the total disintegration rate. The dependence on this decay scheme parameter affects seriously the overall uncertainty due to its large contribution ($\sim$2.5%). The conversion electrons of the 142 keV gamma transition are also measured in the beta counter. Since the internal conversion coefficient for this transition is very high (40.9) their contribution to the beta counter amounts around 10%. For this method, the final accuracy also depends on how well the Compton events below the X-ray peak can be subtracted.

On the other hand, the second method yields the total disintegration rate. However, the detection of 1.6–2.1 keV conversion electrons by the proportional counter is difficult and the efficiency is usually very low, affecting the extrapolation curve results.

Another aspect to be considered is the short half-life which makes difficulties in the beta channel efficiency variation by placing absorbers above and below the radioactive sources. The alternative method is the efficiency variation by electronic discrimination, and this procedure was chosen for the present experiment applying a software coincidence counting system recently installed at the LMN.

For comparison with experimental data, the LMN has developed a methodology for predicting the behavior of extrapolation curves for radionuclide standardization by $4\pi\beta-\gamma$ coincidence

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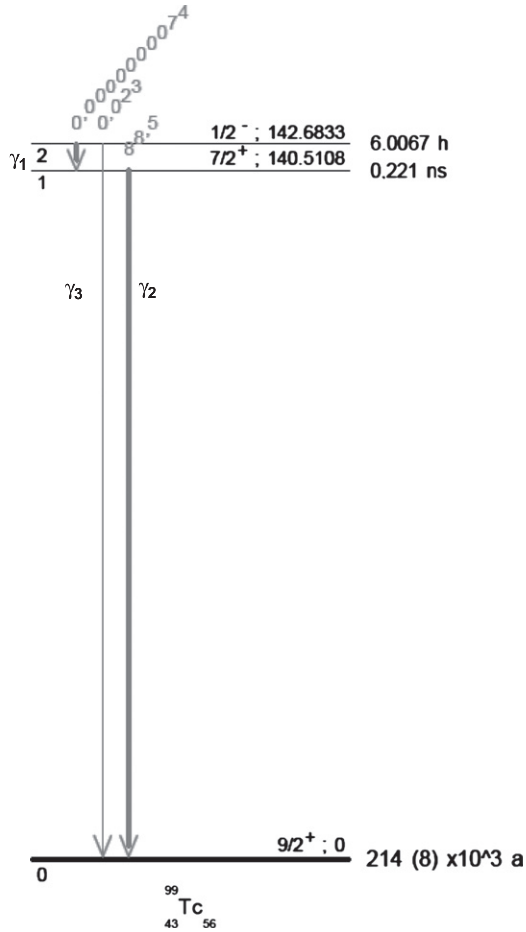

Fig. 1. Decay scheme of ^{99m}Tc (Bé et al., 2004). The energy levels are given in keV. The values above transitions correspond to intensities in percent.

measurements, using the Monte Carlo technique. This methodology has been applied to ^{99m}Tc in order to obtain reliable values for the extrapolation curve. Updated detector response functions were obtained by means of code MCNPX (ORNL, 2006) applied to an improved version of code ESQUEMA (Takeda et al., 2005; Dias et al., 2006). This version includes X-ray detection in both channels coming from conversion electron processes.

2. Methodology

2.1. Coincidence equations

A full description of the coincidence equations can be found elsewhere (Campion, 1959; Baerg, 1966, 1967, 1973). In the case of ^{99m}Tc, these equations are described in the literature (Goodier and Williams, 1966; Ayres and Hirshfeld, 1982; Sahagia, 2006). The general formulae applied to the coincidence measurement were taken from Sahagia (2006) and are given by

$$\frac{N_{4\pi}}{N_0} = a\varepsilon_{ce1} + a(1-\varepsilon_{ce1})\left\{\frac{\alpha_{2T}}{1+\alpha_{2T}}\left[\varepsilon_{ce2}+(1-\varepsilon_{ce2})\varepsilon_{XA(K+L)}\right]+\frac{1}{1+\alpha_{2T}}\varepsilon_{\beta\gamma 2}\right\} + (1-a)\left\{\frac{\alpha_{3T}}{1+\alpha_{3T}}\left[\varepsilon_{ec3}+(1-\varepsilon_{ec3})\varepsilon_{XA(K+L)}\right]+\frac{1}{1+\alpha_{3T}}\varepsilon_{\beta\gamma 3}\right\} \quad (1)$$

$$\frac{N_{\gamma}}{N_0} = a\left(\frac{\alpha_{2K}}{1+\alpha_{2T}}\omega_K\varepsilon_{\gamma XK}+\frac{1}{1+\alpha_{2T}}\varepsilon_{\gamma 2}\right) + (1-a)\left(\frac{\alpha_{3K}}{1+\alpha_{3T}}\omega_K\varepsilon_{\gamma XK}+\frac{1}{1+\alpha_{3T}}\varepsilon_{\gamma 3}\right) \quad (2)$$

$$\frac{N_c}{N_0} = a\left(\varepsilon_{ce1}\frac{1}{1+\alpha_{2T}}\varepsilon_{\gamma 2}+\left[\varepsilon_{ce1}+(1-\varepsilon_{ce1})\varepsilon_{ce2}\right]\frac{\alpha_{2K}}{1+\alpha_{2T}}\omega_K\varepsilon_{\gamma XK}\right) + (1-a)\varepsilon_{ce3}\frac{\alpha_{3K}}{1+\alpha_{3T}}\omega_K\varepsilon_{\gamma XK} \quad (3)$$

where N_0 is the disintegration rate, a is the 140.5 keV transition probability per decay, $N_{4\pi}$ is the proportional counter counting rate, N_γ is the γ-channel counting rate, N_c is the coincidence rate, and ε_γ is the NaI(Tl) crystal efficiency for γ-rays. Since the 142 and 140 keV gamma-rays are quite close in energy, $\varepsilon_{\gamma 1}=\varepsilon_{\gamma 2}=\varepsilon_\gamma$ has been considered, ε_{ce1} is the proportional counter efficiency for (1.73–2.13) keV conversion electrons, $\varepsilon_{X,A}$ is the proportional counter efficiency for X-rays or Auger electrons, ε_{ce2} is the proportional counter efficiency for (119.5–140.5) keV conversion electrons, ε_{ce3} is the proportional counter efficiency for (121.6–142.6) keV conversion electrons, $\varepsilon_{\beta\gamma}$ is the proportional counter efficiency for γ-rays, and α_{iT} is the total conversion coefficient for the ith transition.

In the present paper, the gamma windows were set in two regions: one covering the total energy absorption peak around 20 keV, originating from X-rays, and the other at 140 keV, originating from gamma-rays. These two procedures are called methods 1 and 2.

Combining Eqs. (1)–(3) for the first selected gamma window and assuming the following simplifications:

$\varepsilon_{ce2}=\varepsilon_{ce3}=\varepsilon_{ce}$,
$\varepsilon_{\beta\gamma 2}=\varepsilon_{\beta\gamma 3}=\varepsilon_{\beta\gamma}$ and
$\varepsilon_{\gamma 2}=\varepsilon_{\gamma 3}=0$

It follows that

$$\frac{N_{4\pi}N_\gamma}{N_cN_0} = a\frac{\alpha_{2T}}{1+\alpha_{2T}}+(1-a)\frac{\alpha_{3T}}{1+\alpha_{3T}}+\left(\frac{a}{1+\alpha_{2T}}+\frac{(1-a)}{1+\alpha_{3T}}\right)\varepsilon_{\beta\gamma} + \frac{(1-\varepsilon_{ce})}{\varepsilon_{ce}}\left[\left(a\frac{\alpha_{2T}}{1+\alpha_{2T}}+(1-a)\frac{\alpha_{3T}}{1+\alpha_{3T}}\right)\varepsilon_{XAK} + \left(\frac{a}{1+\alpha_{2T}}+\frac{(1-a)}{1+\alpha_{3T}}\right)\varepsilon_{\beta\gamma}\right] \quad (4)$$

In this case, the proportional counter efficiency is mainly due to 119–140 keV conversion electrons because the lower discriminator level was set above 2.1 keV electron energy. In the extrapolation limit where $(1-\varepsilon_{ce2})\rightarrow 0$, the fraction of N_0 corresponding to the 140 keV + 142 keV conversion electron emission probabilities can be determined. The slope of the extrapolation curve corresponds to the third term in Eq. (4).

Eq. (4) corresponds to the ideal case where the total absorption peaks in the gamma detector can be completely separated. Because of scattering in the NaI(Tl) crystals, part of the 140 keV Compton events falls below the X-ray peak as shown in Fig. 2. This contribution must be subtracted from the X-ray peak in order to keep the simple expression given by (4). These effects can be predicted theoretically by the Monte Carlo method, as explained in Section 2.3.

For the second gamma-ray window, the following simplifications are assumed:

$\varepsilon_{ce2}=\varepsilon_{ce3}=\varepsilon_{ce2}$,
$\varepsilon_{\beta\gamma 2}=\varepsilon_{\beta\gamma 3}=\varepsilon_{\beta\gamma}$ and
$\varepsilon_{XK}=0$

Therefore,

$$\frac{N_{4\pi}N_\gamma}{N_0N_c} = a+(1-a)\left\{\frac{\alpha_{3T}}{1+\alpha_{3T}}\left[\varepsilon_{ce2}+(1-\varepsilon_{ce2})\varepsilon_{X(K+L)}\right]+\frac{1}{1+\alpha_{3T}}\varepsilon_{\beta\gamma}\right\} + \frac{(1-\varepsilon_{ce1})}{\varepsilon_{ce1}}\left\{\left[a\frac{\alpha_{2T}}{1+\alpha_{2T}}+(1-a)\frac{\alpha_{3T}}{1+\alpha_{3T}}\right] \times\left[\varepsilon_{ce2}+(1-\varepsilon_{ce2})\varepsilon_{X(K+L)}\right]+\left[\frac{a}{1+\alpha_{2T}}+\frac{(1-a)}{1+\alpha_{3T}}\right]\varepsilon_{\beta\gamma}\right\} \quad (5)$$

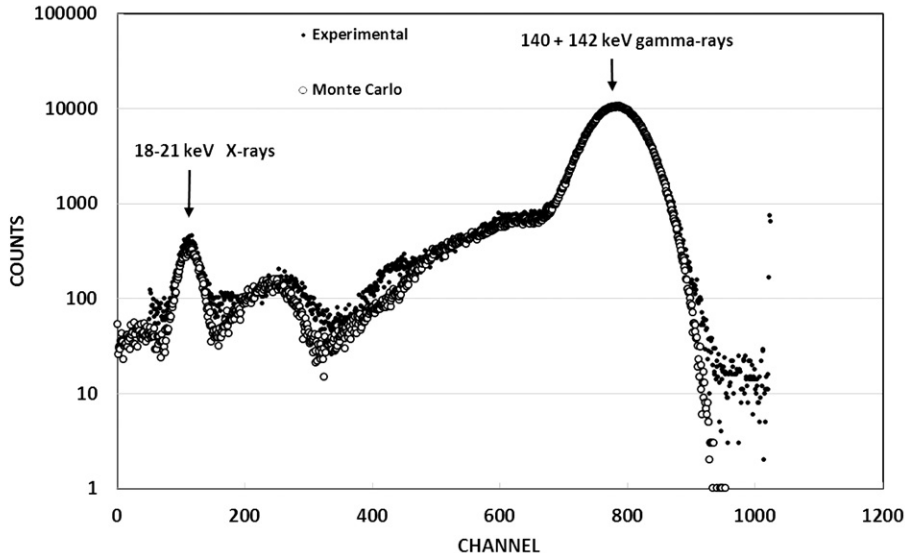

Fig. 2. Gamma-ray spectra obtained for ^{99m}Tc using NaI(Tl) crystal from the $4\pi\beta-\gamma$ system. The black marks correspond to experimental data and white marks to the Monte Carlo calculation using code ESQUEMA (Takeda et al., 2005; Dias et al., 2006, 2010).

In this case, the proportional counter efficiency is mainly due to 1.6–2.1 keV conversion electrons. The terms containing ε_{ce2} in the brackets may be substantially reduced if the upper discriminator level at the 4π detector is set just above the 2.1 keV energy limit. However, some high energy electrons may lose enough energy to fall below this limit. Therefore, some contribution in the beta channel may be due to these high energy electrons. In order to avoid this problem, the upper discriminator level was set to include all deposited electron energies. In this case, all terms inside the bracket in Eq. (5) should be considered. In the extrapolation limit where $(1-\varepsilon_{ce1})\rightarrow 0$, the value of N_0 can be determined.

Corrections for background, dead time and decay were applied to this equation. The corrections of accidental coincidences were performed by means of the Cox-Isham formalism adapted by Smith, (1978).

2.2. Standardization setup

A thin window 4π proportional counter filled with P-10 gas mixture at 0.1 MPa was used as the beta detection channel and a 50.4 mm $\times$ 25.2 mm NaI(Tl) crystal as the gamma detection channel. The software coincidence system was based on a National Instruments PCI-6132 card (National Instruments, 2010) capable of up to four independent analog inputs, and the signals were processed by means of LabView Version 8.5 acquisition program. Information on pulse height and time of occurrence were registered for both beta and gamma channels, together with a third channel corresponding to a reference pulser for checking dead time corrections.

The activity calculation was performed by means of a software coincidence code called SCTAC V.5, developed at the LMN. The pulses reaching the beta channel were produced by 1.6–2.1 keV conversion electrons originated from the 2.17 keV transition, as well as 119–140 keV conversion electrons coming from the 140 keV + 142 keV transitions. The pulses reaching the gamma channel were produced by 18–21 keV X-rays from internal conversion process, as well as gamma-rays from the 140 keV + 142 keV transitions. X-ray detection was possible in the gamma channel due to the presence of a 0.1 mm Al thin window placed at the 4π(PC) detector.

The digital coincidence system made possible two calibration techniques simultaneously: (a) Selecting all electrons above noise at the beta channel and the 140 keV gamma rays in the gamma channel, (b) Selecting electrons above 2.1 keV in the beta channel and 18–21 keV X-rays in the gamma channel. In this case, Compton counts below the X-ray peak have been subtracted.

The ^{99m}Tc solution was obtained by means of at ^{99}Mo generator supplied by the Radiopharmaceutical Center at the IPEN-CNEN/SP followed by *elution* with *physiological* saline *solution*. After this procedure, the radioactive solution was diluted in distilled water and the radioactive sources were prepared by dropping known aliquots on Collodion substrate, previously coated with 10 μg cm^{-2} of gold on both sides. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a desiccator. The accurate source mass determination was performed using a Sartorius MC 21S balance by the pycnometer technique (Campion, 1975).

2.3. Monte Carlo simulation

The theoretical response functions of each detector have been calculated using MCNPX Monte Carlo code (ORNL, 2006). A full description of the $4\pi\beta-\gamma$ system was developed including details of source substrate and absorbers.

Monte Carlo code ESQUEMA (Takeda et al., 2005; Dias et al., 2006, 2010) was developed at LMN and has been used for calculating the extrapolation curve in the $4\pi\beta-\gamma$ coincidence experiment. This code simulates all transitions from the precursor radionuclide to the ground state of the daughter radionuclide, including all detection processes in the coincidence system. The response tables of all detector system components were calculated previously by MCNPX code (ORNL, 2006). As a result, the whole coincidence experiment can be simulated. In this way, Eqs. (4) and (5) could be reproduced by calculation as a function of the beta efficiency, for each of the selected gamma-ray windows yielding the extrapolation curve for each experimental condition. In the present experiment the beta efficiency was varied by pulse height discrimination.

Since X-rays play an important role in ^{99m}Tc simulation by Monte Carlo, improvements in code ESQUEMA have been performed to include X-ray detection produced by conversion electron process in both detector channels. Poisson distribution was considered for the deposited electron energy in order to take into account the finite resolution of the 4π proportional counter.

An X-ray source of ^{54}Mn was used to match the experimental peak broadening in the beta channel.

The final extrapolated value N_0 was obtained by minimizing the following χ^2 equation:

$$\chi^2 = (\vec{y}_{exp} - N_0\vec{y}_{MC})^T V^{-1} (\vec{y}_{exp} - N_0\vec{y}_{MC}) \quad (6)$$

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

A series of simulated values were calculated for a wide range of beta efficiency parameter in small bin intervals. The $\vec{y}_{MC}$ values used in Eq. (6) correspond to the same efficiencies obtained experimentally. This methodology applied co-variance analysis (Smith, 1991) and was compared to linear least square fitting to experimental data.

3. Results and discussion

Fig. 2 shows the theoretical gamma-ray spectrum calculated by code ESQUEMA for ^{99m}Tc, in comparison with the $4\pi\beta-\gamma$ coincidence experiment. The peak to the right corresponds to 140 keV gamma-rays. The small peak to the left corresponds to 18–21 keV and is originated from X-rays emitted from conversion electron process. In this case, the background under the peak is mainly due to Compton events from the 140 keV (or 142 keV) gamma ray and has been subtracted by considering the counts in the neighborhood of the X-ray total absorption peak. The structure in the middle corresponds to Compton or backscattered photons. The black marks correspond to experimental results and white marks correspond to the Monte Carlo calculation. Good agreement can be observed between both spectra.

Fig. 3 shows two 4π detector spectra: in black, the spectrum corresponding to a ^{99m}Tc source deposited on Collodion film previously coated with a 10 μg cm^{-2} thick gold layer on both sides is shown. The contribution from 1.6 to 2.1 keV conversion electrons can be easily seen; the white marks correspond to the same source with three additional 50 μg cm^{-2} Collodion films applied to both sides. It can be noticed that after addition of films these low energy conversion electrons were vanished from the spectrum leaving an approximately flat bottom line due to 119–140 keV conversion electrons.

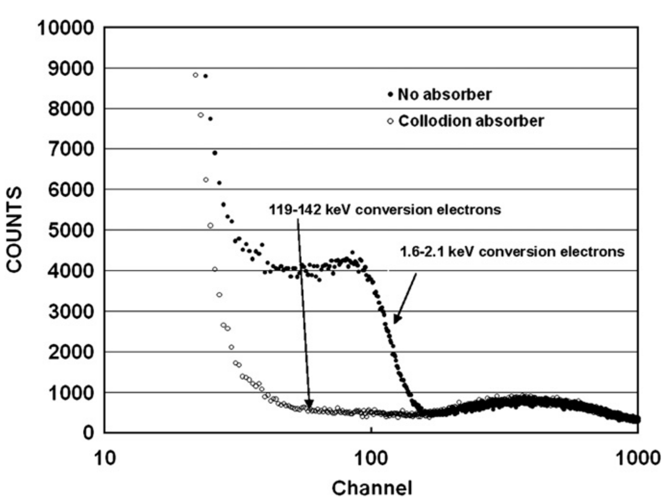

Fig. 3. Spectra of the 4π detector. The black marks correspond to a typical ^{99m}Tc source mounted on Collodion substrate without any additional absorber. The white marks correspond to the same source with three additional 50 μg cm^{-2} thick Collodion films.

Fig. 4 shows the experimental extrapolation curve obtained measuring the whole spectrum above 2.1 keV in the beta channel and setting the gamma channel at 18–21 keV (method 1). In this case, the ^{99m}Tc source was measured without any additional Collodion absorber and the beta efficiency was changed by pulse height discrimination using the *software coincidence counting* system and applying offline analysis with code SCTAC V.5. The Monte Carlo calculation shows a slightly flatter behavior and the maximum calculated efficiency is lower than the experimental one, indicating that the calculation needs improvement for this method. Nevertheless, the agreement between experimental and calculated curves is reasonable within the statistical error in the calculation (~0.5%) for this efficiency range.

It should be pointed out that by shifting the discriminator level in the beta channel both ε_{ce2} and ε_{XAK} are changed. For this reason, the expected behavior of the extrapolation curve is non-linear but

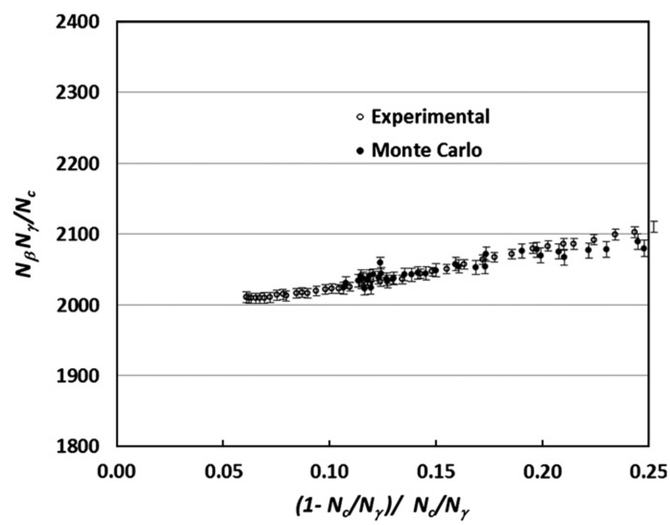

Fig. 4. Extrapolation curves obtained for ^{99m}Tc, gating the gamma channel at 18–21 keV total absorption peak and counting the whole 4π detector spectrum above 2.1 keV. The white marks are experimental points and the black marks are the Monte Carlo calculation.

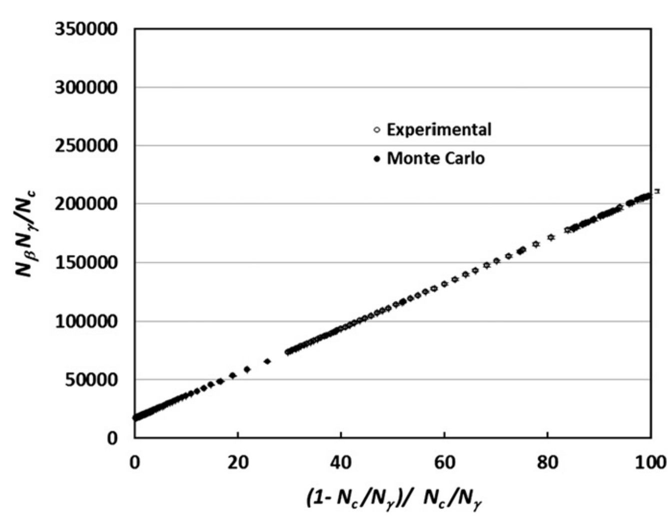

Fig. 5. Extrapolation curves obtained for ^{99m}Tc, gating the gamma channel at 140 keV total absorption peak and counting the whole 4π detector spectrum above a variable lower level pulse height. The white marks are experimental points and the black marks are the Monte Carlo calculation. The error bars in the Monte Carlo results are too small to be visible (<0.5% statistical error).

it is nearly flat in the region of high beta efficiency, as can be seen by Fig. 4.

Fig. 5 shows the experimental extrapolation curve obtained by measuring the whole spectrum in the beta channel and setting the gamma channel at 140 keV total absorption peak (method 2). The beta efficiency was changed the same way as in the previous case. The experimental inefficiency parameter starts around 11.0 because only a small fraction of 1.6–2.1 keV conversion electrons could be detected. The Monte Carlo simulation with code ESQUEMA is shown with white marks. A very good agreement can be observed between the experimental and the theoretical extrapolation curves which are very close to straight lines. The slope is very close to the expected value, given by Eq. (5).

The extrapolation curve parameters of three sources of ^{99m}Tc, prepared from the same batch, obtained with the two methods described in this paper are presented in Table 1. These parameters were obtained by linear least square fitting to experimental data. For method 1 the curve behavior is nearly flat in the region of high beta efficiency. Using the values in this region an average value was obtained by fitting a constant by a least squares technique. For the measurements using this method, it is necessary to apply a correction factor to the extrapolated value in order to determine the activity due to the conversion electron probability per decay. This value is due to the first two terms in Eq. (4) and resulted in 0.114(3). The final weighted average activities are presented in Table 2, compared with Monte Carlo values. The values are in agreement within the experimental uncertainty. The best fit for method 2 was a straight line and the agreement with the Monte Carlo prediction is excellent.

Applying the digital coincidence system it is possible to determine the activity also by selecting only 2 keV electrons at the beta channel and the 140 keV gamma rays in the gamma channel, as suggested by Sahagia (2006). By this method the extrapolation slope is smaller than that of method 2, however this method presents an inconvenience which is the subtraction of the 119 keV conversion electron contribution under the 2 keV spectrum, which is difficult to estimate.

Table 3 shows the partial uncertainties involved in the present experiment. For method 1 the main contribution comes from the value of the conversion electron probability per decay and from the background under the X-ray peak. For method 2, the main contribution comes from the extrapolation procedure.

Table 1
Extrapolation parameters obtained for the two described methods.

Source	Method 1		Method 2
	Intercept[a] (10^3 cps g^{-1})	Intercept[a] (kBq g^{-1})	Slope[a]
1	**11.06 ± 0.03**	**94.88 ± 0.68**	**0.1144 ± 0.0009**
2	**10.91 ± 0.01**	**95.08 ± 0.59**	**0.1124 ± 0.0007**
3	**10.56 ± 0.06**	**95.84 ± 0.60**	**0.1067 ± 0.0007**

[a] Statistical uncertainty.

Table 2
Comparison of activity results obtained from the two methods.

Source	Method 1 (kBq g^{-1})	Method 2 (kBq g^{-1})	Monte Carlo[a] (Method 2) (kBq g^{-1})
1	**97.4 ± 2.8**	**94.88 ± 0.76**	**93.76 ± 0.75**
2	**96.4 ± 2.8**	**95.08 ± 0.68**	**93.96 ± 0.67**
3	**93.0 ± 2.7**	**95.84 ± 0.69**	**94.72 ± 0.68**
Weighted average	**95.6 ± 1.6**	**95.29 ± 0.41**	**94.16 ± 0.41**

[a] Monte Carlo slope = (0.1141 ± 0.0009).

Table 3
Partial uncertainties involved in activity determination (in percent, at 68% confidence level) Types A and B are statistical and systematic errors, respectively.

Component	Type of error	Method 1	Method 2	Remarks
Counting statistics	**A**	**0.25**	**0.63**	**Statistics in extrapolation**
Weighing	**B**	**0.10**	**0.10**	**Balance certificate**
Dead time	**B**	**0.10**	**0.10**	**Pulser method**
Background	**A**	**1.45**	**0.30**	**Statistics counting**
Decay	**B**	**0.02**	**0.02**	**Half-life**
Gandy effect	**B**	**< 0.05**	**< 0.05**	
Resolving time	**B**	**0.10**	**0.10**	**Statistics in accidental coincidence corrections**
Correction factor	**B**	**2.5**	**–**	**Conversion electron probability per decay**
Combined uncertainty		**2.91**	**0.72**	

4. Conclusion

Absolute activity measurements of ^{99m}Tc were carried out by means of a *software coincidence system* developed at the LMN and the results were compared to Monte Carlo simulations. Very good agreement between the experimental and simulated extrapolation curves was observed for the second standardization method indicated by Eq. (5) in which 1.6–2.1 keV conversion electrons are in coincidence with 140.5 keV gamma-rays. Reasonable agreement was obtained between Monte Carlo calculations and experiment for the first method given by Eq. (4), in which 119–140 keV conversion electrons are in coincidence with 18–21 keV X-rays. The experimental efficiency in the beta channel reached a maximum value around 95% whereas in Monte Carlo calculation this value was near 90%. One reason for this difference maybe the resolution function applied to the calculated spectrum in the proportional counter that affects the low energy region of the spectrum. Further improvements are needed in order to achieve better agreement.

The experimental activities obtained for the two methods are in agreement within the experimental uncertainty, indicating that the adopted methodology is adequate.

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