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Standardization of ^{99m}Tc

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Abstract

The radioactivity of ^{99m}Tc was standardized by the 4πPC-γ coincidence method with two different modes. One is using coincidences between (119.5–142.6) keV conversion electrons and *K* X-rays, and the other is coincidences between the 2.13 keV conversion electrons and 140.5 keV γ-rays. The background of the *K* X-ray peak and the sensitivity of the proportional counter (PC) to 140 keV γ-rays were the main sources of uncertainties in the first case and low detection efficiency for conversion electrons in the second case. General coincidence equations were written, with specific forms, for the three measurement variants, including literature variant. Comparison with the ionization chamber calibration is reported.

Keywords: ^{99m}Tc standardization; Coincidence method; Conversion electrons; X-rays

1. Introduction

Absolute standardization of the radionuclide ^{99m}Tc by the coincidence method was reported by Goodier and Williams (1966), who measured coincidences between 2.13 keV conversion electrons and 140.5 keV γ-rays, counting the whole spectrum in the proportional counter (PC). The method was further developed by Ayres and Hirshfeld (1982) by using the same measurement conditions, with an uncertainty of 0.95%.

The present paper proposes two new variants of the coincidence methods for the standardization of ^{99m}Tc; one is based on the use of the coincidences between the (119.5–142.6) keV conversion electrons and the *K* X-rays, with energies of (18.25–21.01) keV, and the other is based on the use of the coincidences between 2.13 keV conversion electrons and 140.5 keV γ-rays, with the main detection of the 2.13 keV conversion electrons in the PC. Comparisons of these two variants as well as previous reported variant are presented.

2. Basic coincidence equations

The decay scheme and parameters as shown in Fig. 1 are taken from BNM-CEA/DAMRI/LNHB "Radionucléides" 1998. If one detects all the emitted radiations, without the *L* X-rays, undetected in the γ-detector, the general counting rates on the 4πPC, NaI(Tl), and coincidence channels are:

$$\frac{N_{4\pi}}{N_0} = a\varepsilon_{ce1} + a(1-\varepsilon_{ce1})\left\{\frac{\alpha_{2T}}{1+\alpha_{2T}}[\varepsilon_{ce2} + (1-\varepsilon_{ce2})\varepsilon_{XA(K+L)}] + \frac{1}{1+\alpha_{2T}}\varepsilon_{\beta\gamma 2}\right\} + (1-a)\left\{\frac{\alpha_{3T}}{1+\alpha_{3T}}\left[\varepsilon_{ce3} + (1-\varepsilon_{ce3})\varepsilon_{XA(K+L)}\right] + \frac{1}{1+\alpha_{3T}}\varepsilon_{\beta\gamma 3}\right\},$$

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

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

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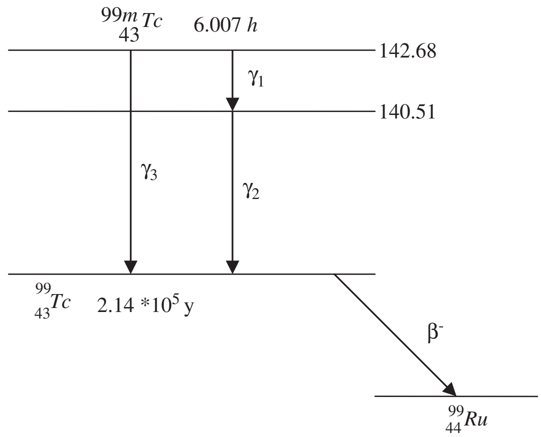

Fig. 1. ^{99m}Tc decay scheme.

where N_0 is the activity, $N_{4\pi}$, N_γ , N_c are counting rates; $a = 0.9912$, is the branching ratio of the 2.17 and 140.5 keV transitions; ε_{ce1} is the efficiency of the PC for the (1.73–2.13) keV conversion electrons from the 2.17 keV transition, while ε_{ce2} and ε_{ce3} are efficiencies for the 119.5–140.5 keV and 121.6–142.6 keV conversion electrons corresponding to the 140.5 and 142.7 keV transitions, respectively: $\varepsilon_{ce2} = \varepsilon_{ce3}$; $\varepsilon_{XA(K+L)}$ is the efficiency of the PC to the L Auger electrons (1.6–2.9) keV, K Auger electrons (14.9–21.0) keV, L X-rays (2.13 keV) and K X-rays (18.25–21.01) keV; $\varepsilon_{\beta\gamma 2}$ and $\varepsilon_{\beta\gamma 3}$ are efficiencies for 140.5 and 142.7 keV γ-rays, respectively: $\varepsilon_{\beta\gamma 2} = \varepsilon_{\beta\gamma 3} = \varepsilon_{\beta\gamma}$; α values are conversion coefficients: $\alpha_{2K} = 0.098$, $\alpha_{2L} = 0.013$; $\alpha_{2T} = 0.114$, $\alpha_{3K} = 29$, $\alpha_{3L} = 9.4$ and $\alpha_{3T} = 41$; $\omega_K = 0.782$ is the fluorescence yield; $\varepsilon_{\gamma XK}$ is the efficiency of the NaI(Tl) detector for K X-rays, $\varepsilon_{\gamma 2}$ and $\varepsilon_{\gamma 3}$ are its efficiencies for 140.5 and 142.7 keV γ-rays, respectively: $\varepsilon_{\gamma 2} = \varepsilon_{\gamma 3} = \varepsilon_\gamma$.

In Eq. (1), coincidences produced by a single photon, scattered in the PC and then registered by the γ-detector were neglected; the reason is the small value of $\varepsilon_{\beta\gamma}$ produced mainly by photo effect. Application of the efficiency extrapolation method allows for the carrying out of the decay scheme corrections. Eq. (1) may be written in various equivalent forms, suited for the application of the method, corresponding to the following various measurement conditions:

(i) Counting of the (119.5–142.6) keV conversion electrons in the PC and of the K X-rays in the γ-channel: $\varepsilon_{ce1} \cong 0, \varepsilon_\gamma \cong 0$, $\varepsilon_{XA(K+L)} = \varepsilon_{XAK}$. Eq. (1) may be written as

$$\frac{N_{4\pi}N_\gamma}{N_c N_0} = 0.11 + 0.89\varepsilon_{\beta\gamma} + \left(\frac{1-\varepsilon_{ce2}}{\varepsilon_{ce2}}\right)(0.093\varepsilon_{XAK} + 0.89\varepsilon_{\beta\gamma}), \tag{2}$$

with

$$\frac{N_c}{N_\gamma} \cong \varepsilon_{ce2}. \tag{3}$$

(ii) Counting of the (1.73–2.13) keV conversion electrons in the PC and of the γ-rays. In this case, $\varepsilon_{ce2} \cong 0$, $\varepsilon_{\gamma XY} = 0$, $\varepsilon_{XA(K+L)} = \varepsilon_{XAL}$ and Eq. (1) become

$$\frac{N_{4\pi}N_\gamma}{N_c N_0} = 0.9912 + 0.0001\varepsilon_{XAL} + 0.0002\varepsilon_{\beta\gamma} + \left(\frac{1-\varepsilon_{ce1}}{\varepsilon_{ce1}}\right)(0.0135\varepsilon_{XAL} + 0.89\varepsilon_{\beta\gamma}) \tag{4}$$

with

$$\frac{N_c}{N_\gamma} \cong \varepsilon_{ce1}. \tag{5}$$

(iii) The measurement conditions used by Goodier and Ayres: counting of the whole spectrum of radiations detected in the PC and counting of the γ-rays. In this case, $\varepsilon_{ce2} = \varepsilon_{ce3} \cong 1$, $\varepsilon_{\gamma XK} = 0$ and Eq. (1) become

$$\frac{N_{4\pi}N_\gamma}{N_c N_0} = 1 + 0.0002\varepsilon_{\beta\gamma} + \left(\frac{1-\varepsilon_{ce1}}{\varepsilon_{ce1}}\right)(0.11 + 0.89\varepsilon_{\beta\gamma}), \tag{6}$$

with

$$\frac{N_c}{N_\gamma} \cong \varepsilon_{ce1}. \tag{7}$$

The above equations emphasize the following aspects:

Variant (i) has the main advantage of a high efficiency ε_{ce2} and consequently of a short extrapolation interval. The main problem in this application is to remove a significant contribution of the Compton of 140 keV γ-rays. Another correction is due to $\varepsilon_{\beta\gamma}$ which must be determined carefully; the extrapolation may be nonlinear, due to the variation of the ε_{XAK} during the variation of ε_{ce2}.

Variant (ii) has the main advantage of a small extrapolation slope. The main disadvantage is the low value of ε_{ce1}, and consequently, a large extrapolation interval; it introduces also a supplementary correction of the extrapolated value, which is negligible, as the coefficients of the small values ε_{XAL} and is $\varepsilon_{\beta\gamma}$ are negligible.

Variant (iii) is advantageous due to the simplicity, and due to the high counting rates, but the value of the efficiency ε_{ce1} is also low and the extrapolation interval is large. The extrapolation slope is high, that introduces a supplementary uncertainty.

3. Experimental procedure

3.1. Preparation of sources

The sources were prepared gravimetrically from three different Na[^{99m}Tc]O$_4$ solutions, which were first measured in the calibrated ionization chamber of IFIN. A set of two sources were prepared from an original solution, containing 0.9% NaCl which resulted in a relatively high self-absorption. The third source was prepared from diluted solution by a factor of $F = 17.881$ with distilled water.