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Absolute Disintegration Rate Measurements of Ga-67

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The procedure for determining the ^{67}Ga disintegration rate by means of a $4\pi\beta$–γ coincidence system is described. The main difficulty with this radionuclide is the presence of a meta-stable state with a half-life of 9.1 μs and a total internal conversion coefficient of 0.89. This required several measurements using different electronic dead-times to correct for detection of delayed events. Gamma spectrometry measurements by a HPGe detector were also performed in order to check for impurities. © 1998 Elsevier Science Ltd. All rights reserved

Introduction

One of the consequences of the production of new radioisotopes for nuclear medicine, such as ^{67}Ga, is the need for standard sources of these radionuclides to be obtained in a fast and simple way. The Laboratório de Metrologia de Radionuclídeos (LMR) at IPEN in São Paulo, has a well-type ionization chamber system, which is ideal for this purpose. In order to calibrate this system it was necessary to standardize ^{67}Ga solutions by an absolute method.

The decay scheme of ^{67}Ga is shown in Fig. 1 (Lagoutine *et al.*, 1984). This nuclide decays with a half-life of 3.259 d by electron capture, populating the excited levels of ^{67}Zn. The internal conversion coefficients are generally small, except for the 93.3-keV transition of ^{67}Zn, which has a meta-stable state with 9.1 μs half-life and a total internal conversion coefficient of 0.89.

The measurements were carried out in a $4\pi\beta$–γ coincidence system (Moura, 1969; Fonseca, 1997) as described in Section 2. The activity has been determined by making use of the efficiency extrapolation method (Baerg, 1973) The dead-time set by the electronic system was varied and the results were extrapolated to infinite dead-time, in order to correct for delayed events as described in Sections 2 and 3.

Experimental

Source preparation

The ^{67}Ga solution was obtained from the batches routinely imported from Nordion (Canada) and processed by IPEN as they would be delivered for medical applications. The radioactive solution consisted of gallium citrate diluted in 0.16 N HCl solution. The counting sources, subsequently, were prepared by dispensing known aliquots of the solution on 20 μg/cm^2 thick Collodion films. This film had been previously coated with a 10 μg/cm^2 thick gold layer in order to render the film conductive. A seeding agent (Cyastat SN) was used for improving the deposit uniformity and the sources were dried in a warm (45°C) nitrogen jet (Wyllie *et al.*, 1970). The accurate source mass determination was performed using the pycnometer technique (Campion, 1975).

$4\pi\beta$–γ coincidence measurements

The system for the absolute standardization of the ^{67}Ga sources consisted of a 4π gas-flow proportional counter coupled to a pair of 3 × 3 inch NaI(Tl) scintillation counters, operating in coincidence. The proportional counter was used to detect Auger electrons and X-rays from electron capture events, as well as prompt and delayed internal conversion electrons from the excited-state transitions. The measurements were made with radioactive solutions from different batches and were normalized by measuring each of them in a γ-spectrometer. This normalization was performed by measuring the 185-keV photopeak areas of ^{67}Ga with a HPGe

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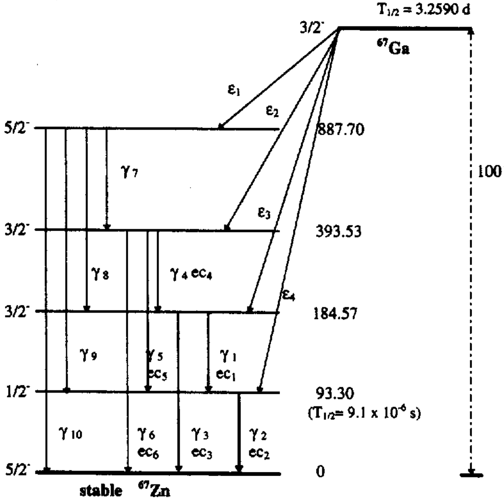

Fig. 1. Decay scheme of ^{67}Ga (Lagoutine *et al.*, 1984). The energies are in keV.

spectrometer for different batches, and taking the ratios with respect to the first batch.

The number of detected events in the proportional counter N_X is given by:

$$N_X = N_0 \sum_r a_r \left\{ \varepsilon_X + \left(\frac{\alpha_r \varepsilon_{ce}}{(1+\alpha_r)} \right)_{\text{delayed}} e^{-\lambda\tau} + (1 - \varepsilon_X) \left[\left(\frac{\alpha_r \varepsilon_{ce} + \varepsilon_{\beta\gamma}}{(1+\alpha_r)} \right)_{\text{prompt}} + \left(\frac{\alpha_r \varepsilon_{ce} + \varepsilon_{\beta\gamma}}{(1+\alpha_r)} \right)_{\text{delayed}} \right] \right\} \tag{1}$$

In (1), N_0 refers to the source activity, coefficient a_r is the branching ratio of energy level r, ε_X is the detection efficiency for X-rays and Auger electrons, ε_{ce} is the detection efficiency of conversion electrons, α_r is the total conversion coefficient for branch r and $\varepsilon_{\gamma 3}$ is the gamma efficiency of gamma 3. The parameter $\varepsilon_{\beta\gamma}$ is the proportional counter efficiency for γ rays.

For the γ-channel a window was set in the energy peak of 185 keV, which is the most intense γ and is not in coincidence with the 93.3-keV transition and the number of registered events N_γ is given by

$$N_\gamma = N_0 a_3 \varepsilon_{\gamma 3} \tag{2}$$

The number of coincident events is therefore given by

$$N_C = N_0 a_3 \varepsilon_X \varepsilon_{\gamma 3}. \tag{3}$$

The values of N_X and N_γ have been corrected for background, dead-time and decay. The value of N_C was corrected for dead-time and accidental coincidences applying the Cox–Isham formalism (Cox and Isham, 1977).

The ratio of (3) and (2) gives the efficiency parameter ε_X and (1)–(3) lead to the activity value by the expression:

$$\frac{N_X N_\gamma}{N_C} = N_0 \left(1 + \left(\frac{\alpha_r \varepsilon_{ce}}{(1+\alpha_r)} \right)_{delayed} e^{-\lambda\tau} + k_\varepsilon \right), \tag{4}$$

where $k_\varepsilon = [(1 - (N_C/N_\gamma))/(N_C/N_\gamma)]C$, $C = [(\alpha_r \varepsilon_{ce} + \varepsilon_{\beta\gamma})/(1 + \alpha_r)]_{\text{prompt}} + [(\alpha_r \varepsilon_{ce} + \varepsilon_{\beta\gamma})/(1 + \alpha_r)]_{\text{delayed}}$, λ is the decay constant of the 93.3-keV meta-stable level and τ is the dead-time defined electronically.

The effects of the delayed events were corrected by extrapolating the activity to infinite dead-time. For this extrapolation, a series of measurements using dead-times of 3, 5, 10, 13, 19, 24, 28 and 48 μs have been performed. The dead-times (non-extending type) were measured with an uncertainty of ±0.02 μs, by means of the source-pulser method (Baerg, 1965). The extrapolation to infinite dead-time has been performed by considering the following expression:

$$f_\tau = \frac{A_0 P_D e^{-\lambda\tau}}{1 + P_D e^{-\lambda\tau_3}}, \tag{5}$$

where $P_D = (\alpha_r/(1+\alpha_r))_{\text{delayed}}$ and τ is the electronic dead-time. P_D is the emission probability for delayed conversion electrons and has been obtained experimentally by a preliminary fitting of $N_X N_\gamma / N_C$ as a function of $(1 - N_C/N_\gamma)/N_C/N_\gamma$ for a fixed dead-time (τ_3) of 2.899 μs. The slope of the curve is given by $(P_D + P_P)$ where P_P is the contribution from prompt conversion electrons to the proportional counter response. The intercept of this preliminary curve is A_0, given by

$$A_0 = N_0 (1 + P_D e^{-\lambda\tau_3}). \tag{6}$$

Since the contribution of P_P is small, it has been subtracted from the experimental slope by using the ratio P_P/P_D from the literature (Lagoutine *et al.*, 1984). At each dead-time, the value of f_τ has been subtracted from the quantity $N_X N_\gamma / N_C$. In this way all values of $N_X N_\gamma / N_C$ obtained at different dead-times are corrected for delayed events. The slope of the corrected curve of $N_X N_\gamma / N_C$ as a function of $(1 - N_C/N_\gamma)/N_C/N_\gamma$ corresponds to a new value of $(P_D + P_P)$. This value is applied again in (5) in a iterative procedure until a converging value of $(P_D + P_P)$ is achieved. The intercept of the final curve is the activity N_0. This procedure is equivalent to extrapolating the observed values of $N_X N_\gamma / N_C$ to infinite dead-time because the intercept does not depend on delayed events.

The meta-stable half-life has been measured by feeding the signals from the proportional counter to a time to amplitude converter, previously calibrated by means of an accurate time mark generator.

Results and Discussion

Figure 2 shows the behavior of $N_X N_\gamma / N_C$ as a function of $(1 - N_\gamma/N_C)/(N_\gamma/N_C)$. In these curves ε_X

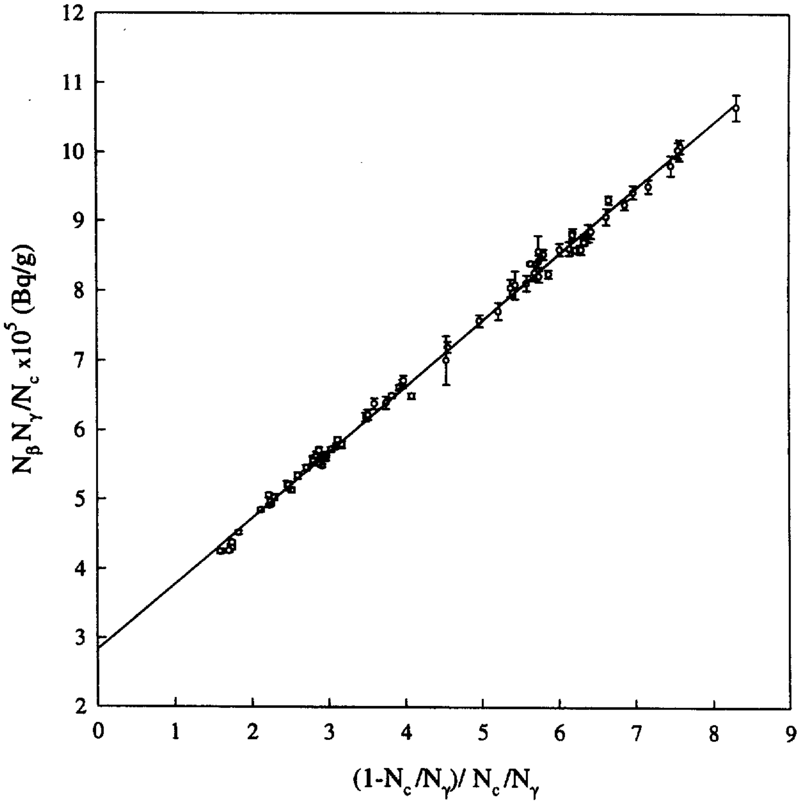

Fig. 2. Extrapolation curve of $N_X N_\gamma/N_C$ as a function of $(1 - N_\gamma/N_C)/(N_\gamma/N_C)$.

was varied in the range of 15 to 46%. All data taken with different dead-times are shown in Fig. 2. It can be seen that, apart from some fluctuations, most points follow a linear behavior. The slope of this curve $(33.6 \pm 0.4)\%$ corresponds to $(P_D + P_D)$ as compared to the values from the literature $(35.49 \pm 0.60)\%$ (Lagoutine *et al.*, 1984) and $(31.9 \pm 0.9)\%$ (Browne and Firestone, 1986). In this curve, the intercept corresponds to the corrected activity N_0. The uncertainty in this value was 1.1%. The measured half-life of (9.34 ± 0.20) μs agreed with the value from the literature of 9.1 μs (Lagoutine *et al.*, 1984).

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