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Standardization of ^{67}Cu and calibration of the ionization chamber. Impurities and decay scheme problems

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ABSTRACT

This paper describes the standardization of ^{67}Cu by the 4πβ(PC)-γ coincidence method, and the calibration of ionization chamber. The difficulties were. (i) One of the excited levels of ^{67}Zn has a half life of 9.10 μs. (ii) A recent publication reconsiders the decay scheme parameters. (iii) The solution had a significant content of impurities. The conclusions were that the result of absolute standardization is less influenced by the decay scheme parameters and impurities than the measurements by ionization chamber.

HIGHLIGHTS

- Standardization by the 4πβ(PC)-γ coincidence method.
- Calibration of the ionization chamber.
- Problems: Excited level of ^{67}Zn, decay scheme, impurity.
- Absolute standardization is less influenced than the ionization chamber measurement.

1. Introduction

^{67}Cu is a beta-gamma emitter with a half life of 61.83 h, with a complex decay scheme of the triangular type, resulting in the rather weak emission of gamma-rays with low energy, mainly 184.6 keV, intensity 48.7%, and emission of beta minus radiations, maximum energy within the interval (168.2–561.7) keV, intensity 100%. It is of great interest in targeted radiotherapy, especially radioimmunotherapy, due to the properties: (a) It can be associated in the same type of pharmaceutical with ^{61}Cu, $T_{1/2}$ = 3.33 h, a positron emitter, or with ^{64}Cu, $T_{1/2}$ = 12.701 h, a positron and electron emitter, resulting theranostics products (PET diagnosis + therapy) (Verel et al., 2005); (b) It is superior as compared with ^{131}I, due to its high tropism for some organs and due to its lower energy and emission intensity of gamma-rays, with the result the optimum irradiation of the target treatment volume and avoidance of nonuseful irradiation of the adjacent organs. (DeNardo et al., 1999). It can be produced in several modes (Qaim, 2015; Medvedev et al., 2012) at a cyclotron via the reactions: ^{68}Zn (p, 2p)^{67}Cu; ^{67}Zn (d, 2p) ^{67}Cu; ^{64}Ni (α, p) ^{67}Cu, and at a nuclear reactor via the ^{67}Zn (n, p)^{67}Cu reaction; the common problem is the occurrence of impurities, hard to be removed (Asabella et al., 2014). It can be also produced at the linear accelerator via the reaction ^{68}Zn (γ, p)^{67}Cu, with a high degree of purity, Chen et al. (2015). This paper describes the absolute standardization of ^{67}Cu by the 4πβ(PC)-γ coincidence method. The radioactive solution was obtained from the Nuclear Physics Institute of the ASCR, v.v.i, Department of radiopharmaceuticals, 250 68 Husinec-Rež 130, Czech Republic, by the kindness of Dr. Eng. Ondřej Lebeda. A content of ^{67}Ga of (3.59 ± 0.91)% was detected, a nonnegligible quantity, for which we present the mode to treat its contribution in activity and its subtraction. On the other hand, the problems of uncertainties in decay data, according to new published data, is treated. Finally, the calibration of CENTRONIC IG12/20 A ionization chamber results are presented.

2. Standardization of ^{67}Cu by the 4πβ(PC)-γ coincidence method

2.1. Decay scheme and basic coincidence equations

2.1.1. Neglecting the dead time influence on the counting rate

Fig. 1 presents the decay scheme, from Junde et al. (2005) and (Meyer et al., 1978), with the following intensities and energies of β⁻ emissions: b_1 = 0.011(11), $E_{\beta max}$ = 168.2 keV; b_2 = 0. 57(6), $E_{\beta max}$ = 377.1 keV;

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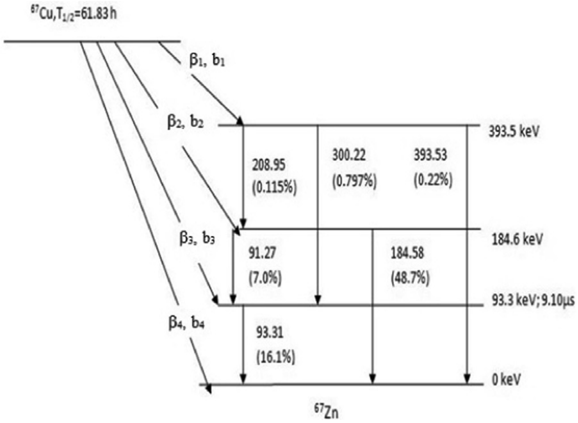

Fig. 1. Decay scheme of ^{67}Cu.

b_3 = 0.220(22), $E_{\beta max}$ = 468.4 keV; b_4 = 0.200(20), $E_{\beta max}$ = 561.7 keV. The low energy transitions towards the ground state of ^{67}Zn are accompanied by internal conversion, as follows. In the transitions from the levels: 393.5 keV and 184.6 keV, low intensity conversion electrons (*ce*), coincident with the respective beta radiations, are emitted. In the isomer transition from the 93.31 keV level, with a half life of 9.10 μs, the ce_3 emissions are E_{cK} = 83.652 keV, [I = 0.1209(15)] E_{cL} = 92.117 keV, [I = 0.01481(18)] and E_{cM} = 93.17 keV, [I = 0.0021], noncoincident with the beta rays. The β_1-radiations are coincident with the (209.0 + 300.2 + 393.5 keV) γ-rays, the β_2 are coincident with the (91.3 and 184.6 keV) ones. The β_3 radiations have not coincident γ-rays and the β_4 are decays to the ground state. The ce_3 are superimposed on the noncoincident β_3. The 93.31 keV γ-rays are not coincident with all the beta radiations feeding the 93.31 keV level, (b_1, b_2 and b_3) and consequently they must be avoided in the counting, by applying a higher energy threshold, let's say 130 keV. Otherwise, a false, lower beta counting efficiency would occur. In this case, the decay scheme can be replaced with the equivalent one, presented in Fig. 2, and the corresponding coincidence equations are:

$$\frac{N_\beta}{N_0} = a_1[\varepsilon_{\beta 1} + (1-\varepsilon_{\beta 1})\frac{\varepsilon_{ce1}\alpha_1 + \varepsilon_{\beta\gamma 1}}{1+\alpha_1}] + a_2[\varepsilon_{\beta 2} + (1-\varepsilon_{\beta 2})\frac{\varepsilon_{ce2}\alpha_2 + \varepsilon_{\beta\gamma 2}}{1+\alpha_2}] +$$
$$(a_3\varepsilon_{\beta 3} + a_4\varepsilon_{\beta 4}) + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}$$
$$\frac{N_\gamma}{N_0} = a_1[\frac{0.00115}{a_1}(\varepsilon_{\gamma 208} + \varepsilon_{\gamma 185}) + \frac{0.00797}{a_1}\varepsilon_{\gamma 300} + \frac{0.0022}{a_1}\varepsilon_{\gamma 393}] + a_2\varepsilon_{\gamma 185}$$
$$= a_1\varepsilon_{\gamma 1} + a_2\varepsilon_{\gamma 2}$$
$$\frac{N_c}{N_0} = a_1\varepsilon_{\gamma 1}[\varepsilon_{\beta 1} + (1-\varepsilon_{\beta 1})\frac{\varepsilon_{c1}}{\varepsilon_{\gamma 1}}] + a_2\varepsilon_{\gamma 2}[\varepsilon_{\beta 2} + (1-\varepsilon_{\beta 2})\frac{\varepsilon_{c2}}{\varepsilon_{\gamma 2}}] \quad (1)$$

ε_β and $\varepsilon_{\beta\gamma}$ are the proportional counter (PC) efficiencies to β radiations and γ-rays. ε_γ are the efficiencies of the NaI(Tl) crystal to γ-rays and ε_c are Compton scattering and γ-γ coincidences. The symbols of α with suffix ce_1 and ce_2 correspond to the weighted means of the internal conversion coefficients for transitions from the 393.5 kev-level and from the 184.6 kev-level, respectively, where the weights are the transition probabilities; ce_3 are the noncoincident 83.652 keV, 92.117 keV and 93.17 keV conversion electrons; ce_4 are concurrent with the non counted 91.27 γ-rays and may be considered as an "electron emission subsequent to the beta radiation" (Grigorescu et al., 2002). α are conversion coefficients corresponding to the respective decay levels.

The linearity condition for branches a_1 and a_2 is: $\varepsilon_{\gamma 1} = \varepsilon_{\gamma 2}$

$$\frac{1}{0.011}[0.00115(\varepsilon_{\gamma 208} + \varepsilon_{\gamma 185}) + 0.00797\varepsilon_{\gamma 300} + 0.0022\varepsilon_{\gamma 393}] = \varepsilon_{\gamma 185} \quad (2)$$

Or approximately: $$0.725\varepsilon_{\gamma 300} + 0.20\varepsilon_{\gamma 393} = 0.791\varepsilon_{\gamma 185} \quad (3)$$

Condition (3) is almost accomplished by imposing the γ counting window between 100 keV and 400 keV. In this situation, a new simplified, equivalent decay scheme can be considered, as presented in Fig. 3. The coincidence relations (1) reduce to:

$$\frac{N_\beta}{N_0} = (a_1 + a_2)[\varepsilon_{\beta mean,\ 1+2} + (1-\varepsilon_{\beta mean,\ 1+2})\frac{\varepsilon_{ce2}\alpha_2 + \varepsilon_{\beta\gamma 2}}{1+\alpha_2}] +$$
$$(a_3 + a_4)\varepsilon_{\beta mean,3+4} + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}$$
$$\frac{N_\gamma}{N_0} = (a_1 + a_2)\varepsilon_\gamma$$
$$\frac{N_c}{N_0} = (a_1 + a_2)\varepsilon_\gamma[\varepsilon_{\beta mean,\ 1+2} + (1-\varepsilon_{\beta mean,\ 1+2})\frac{\varepsilon_c}{\varepsilon_\gamma}] \quad (4)$$

In the linearity conditions, Fig. 3, and for short extrapolation intervals, one may consider a linear efficiency relation (Sahagia et al., 2002), of the type

$$(1-\varepsilon_{\beta mean,3+4}) = m(1 - N_c/N_\gamma) \quad (5)$$

Eq. (4) become:

$$\frac{N_\beta N_\gamma}{N_0 N_c} = 1 + (1-K_1)(\frac{N_\gamma}{N_c} - 1) + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}\frac{N_\gamma}{N_c}$$
$$= 1 + (1-K)(\frac{N_\gamma}{N_c} - 1) + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3};$$
$$(1-K) = (1 - K_1 + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}) \quad (6)$$

When the extrapolation procedure is applied, N_γ/N_c = 1, Eq. (6) becomes:

$$N_0 = (\frac{N_\beta N_\gamma}{N_c})_{extrapolated}\frac{1}{1 + I_{transition\ 93.31\mathrm{keV}}\frac{\varepsilon_{ce3}\alpha_3 + \varepsilon_{\beta\gamma 3}}{1+\alpha_3}} \quad (7)$$

2.1.2. Dead time correction due to the delay of the 93.31 keV state

Due to the significant delay of the 93.31 keV level, $T_{1/2}$ = 9.10 μs, and of the nonnegligible dead time of the beta channel in our coincidence installation, θ = (10.0 ± 0.5) μs, a significant loss of signals

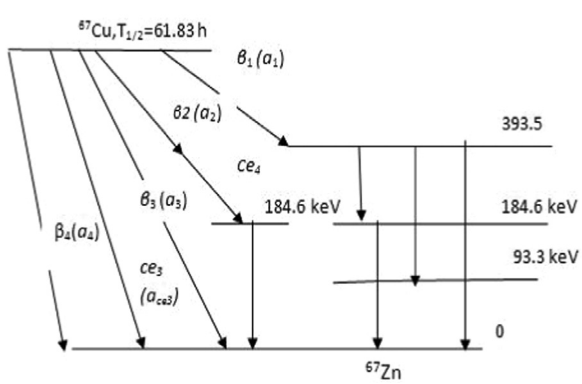

Fig. 2. Equivalent decay scheme of ^{67}Cu.

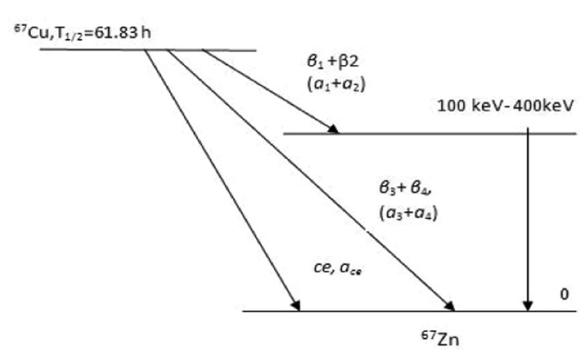

Fig. 3. Simplified equivalent decay scheme of ^{67}Cu.