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OPTIMIZATION OF SAMPLE PREPARATION FOR $4\pi\beta-\gamma$ COINCIDENCE MEASUREMENTS

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ABSTRACT

The influence of different parameters related to radioactive source substrate and preparation techniques to be used in radionuclide standardization, by means of $4\pi(PC)\beta-\gamma$ coincidence systems were studied. These parameters included: Collodion film thickness, gold deposit thickness (on each side), radioactive source mass, seeding agent and aliquot evaporation atmosphere (dessecator or warm nitrogen jet). With this procedure it was possible to verify the influence of each parameter and establish the best conditions for sample preparation.

1. INTRODUCTION

The present work was intended to verify the influence of different parameters related to radioactive source substrate and preparation techniques to be used in radionuclide standardization, by means of $4\pi(PC)\beta-\gamma$ coincidence systems installed in the Nuclear Metrology Laboratory, IPEN, São Paulo.

This system is composed of a proportional counter in 4π geometry for alpha or electron detection, coupled to NaI(Tl) scintillator or HPGe semiconductor detectors for gamma ray detection. The substrate is composed of Collodion film placed on a stainless steel holder. Vacuum evaporated gold substrate is deposited on one or both faces of the film in order to render it conductive.

Several parameters involved were changed, namely: Collodion film thickness, gold deposit thickness (on each side), radioactive source mass, seeding agent (Cyastat) and aliquot evaporation atmosphere (dessecator or warm nitrogen jet). With this procedure it was possible to verify the influence of each parameter on the beta detection efficiency and establish the best conditions for sample preparation.

2. METHODOLOGY

The first part describes the theoretical background involved in coincidence measurements and parameter analysis, followed by a description of the experimental procedure.

2.1. Coincidence Equations

The general coincidence equations applied to β–γ radionuclides are :

$$N_\beta = N_0 \sum_{i=1}^{m} a_i \left\{ \varepsilon_{\beta_i} + \left(1-\varepsilon_{\beta_i}\right) \sum_{j=1}^{n} b_{ij} \frac{\alpha_{ij} \left\lfloor \varepsilon_{CE_{ij}} + (1-\varepsilon_{CE_{ij}})\varepsilon_{(X,A)_{ij}} \right\rfloor + \varepsilon_{\beta\gamma_{ij}}}{1+\alpha_{ij}} \right\} \quad (1)$$

$$N_\gamma = N_0 \sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} \quad (2)$$

$$N_c = N_0 \sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + \left(1-\varepsilon_{\beta_i}\right) \sum_{j=1}^{n} b_{ij}\, \varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right] \quad (3)$$

Therefore,

$$\frac{N_\beta N_\gamma}{N_c} = N_0 \frac{\sum_{i=1}^{m} a_i \left\{ \varepsilon_{\beta_i} + \left(1-\varepsilon_{\beta_i}\right) \sum_{j=1}^{n} b_{ij} \frac{\alpha_{ij} \left\lfloor \varepsilon_{CE_{ij}} + (1-\varepsilon_{CE_{ij}})\varepsilon_{(X,A)_{ij}} \right\rfloor + \varepsilon_{\beta\gamma_{ij}}}{1+\alpha_{ij}} \right\} \sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}}}{\sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + \left(1-\varepsilon_{\beta_i}\right) \sum_{j=1}^{n} b_{ij}\, \varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right]} \quad (4)$$

where:

N_β, N_γ and N_c are beta, gamma and coincidence counting rates, respectively;
N_0 is the radioactive source disintegration rate;
a_i and b_{ij} are the intensity per decay of the i-th beta transition and relative intensity of the j-th transition with respect to the i-th transition;
n is the number of daughter transitions following the i-th beta transition;
m is the number of beta transitions;
$\varepsilon_{\beta i}$ is the beta efficiency associated to i-th beta transition;
$\varepsilon_{\gamma_{ij}}$, and $\varepsilon_{\beta\gamma_{ij}}$ are gamma detection efficiency and gamma efficiency of beta detector, respectively, associated to ij-th transition;
ε_{CEij} and $\varepsilon_{(X,A)ij}$ are conversion electron detection efficiency and electron Auger or X-ray detection efficiency, respectively, associated to ij-th transition, and
ε_{CEij} and α_{ij} are the gamma-gamma coincidence detection efficiency and total internal conversion coefficient of the ij-th transition.

A measure of the beta efficiency can be given by:

$$\frac{N_c}{N_\gamma} = \frac{\sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + \left(1-\varepsilon_{\beta_i}\right) \sum_{j=1}^{n} b_{ij}\, \varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right]}{\sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\, \varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}}} \quad (5)$$

In the special cases where the total energy absorption peak in the gamma channel can be selected as window for a single gamma-ray line, gamma-gamma coincidences are eliminated and equations (4) and (5) can be simplified:

$$\frac{N_\beta N_\gamma}{N_c} = N_0\left\{1 + b\,\frac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\,\frac{\alpha[\varepsilon_{CE} + (1-\varepsilon_{CE})\varepsilon_{(X,A)}] + \varepsilon_{\beta\gamma}}{1+\alpha}\right\} \tag{6}$$

and

$$\frac{N_c}{N_\gamma} = \varepsilon_\beta \tag{7}$$

where $\varepsilon_\beta = \sum_{i=1}^{m} a_i \varepsilon_{\beta i}$

In the present work the beta efficiency was measured as a function of sample preparation parameters for ^{60}Co. This radionuclide has been selected because of simple decay characteristics and low conversion electron coefficient. In this case, the dependence between parameters $N_\beta N_\gamma / N_c$ and N_c/N_γ is small. Furthermore, its beta endpoint energy is sufficiently low (316 keV) yielding a beta efficiency which is sensitive to sample preparation procedures.

2.2. 2.2. Analysis of sample preparation variables

A multi-parametric fitting between the beta efficiency for ^{60}Co obtained from $4\pi(PC)\beta-\gamma$ coincidence measurement and the sample preparation parameters was performed. The relationship is given by:

$$\varepsilon_\beta = \sum_{i=0}^{5} a_i x_i \tag{8}$$

where ε_β is the beta efficiency and x_i are the sample preparation variables.

2.3. Experimental procedure

The standardization of ^{60}Co was performed by means of a conventional $4\pi(PC)\beta-\gamma$ coincidence system, consisting of 4π proportional counter filled with P-10 gas mixture at 0.1 MPa, coupled to a pair of 3” x 3” NaI(Tl) crystals. A detailed description of this system is given elsewhere [1]. All pulses above noise threshold (0.7 keV) were measured in the beta channel. The gamma-rays were gated at a discrimination window covering the total absorption peaks of 1173 keV and 1332 keV, which are not completely resolved by NaI(Tl) scintillator.

The radioactive sources to be measured in the $4\pi(PC)\beta-\gamma$ system were prepared by dropping known aliquots of the ^{60}Co solution on 9-21 $\mu g.cm^{-2}$ thick Collodion films. These film were previously coated with a 6,9-33,1 $\mu g.cm^{-2}$ thick gold layer on one or both sides, in order to render the film conductive. A seeding agent (Cyastat) was used for improving the deposit uniformity and the sources were dried in warm (40 C) nitrogen jet flow. The accurate source mass determination was performed using a Sartorius MC21S electronic balance by means of the pycnometre technique [2].

The variation in the efficiency was achieved by placing external Collodion or aluminum absorbers over or under the radioactive sources. The parameters changed in order to verify the influence on the beta efficiency were: Collodion film thickness, gold deposit thickness (on each side), radioactive source mass, aliquot evaporation atmosphere (0 for dessecator and 1 for warm nitrogen jet) and the seeding agent (0 - no seeding agent; 1 - Cyastat seeding agent). Table 1 shows the sample preparation variables obtained in this experiment.

Table 1 Sample preparation variables obtained in the experiment.

Souce No.	Beta Efficiency	Collodion film thickness ($\mu g/cm^2$)	Evaporation atmosphere 0-dessecat. 1-nitrogen	Seeding agent (Cyastat) 1 - with	Gold deposit thickness Side 1 ($\mu g/cm^2$)	Gold deposit thickness Side 2 ($\mu g/cm^2$)	Radioactive source mass (mg)
12601	82.98	13.3	0	1	0.0	0.0	18.020
12602	87.23	13.3	1	1	0.0	0.0	8.932
12604	87.83	11.4	0	1	16.8	0.0	28.418
12605	90.90	13.3	1	1	17.1	0.0	28.064
12606	86.94	13.1	0	1	15.6	0.0	18.738
12610	86.71	9.4	1	1	0.0	0.0	6.403
12612	85.75	20.5	1	1	0.0	0.0	8.041
12615	88.30	20.9	1	1	15.5	0.0	7.031
12617	90.83	11.1	1	1	12.5	13.8	7.538
12925	81.94	13.6	0	0	8.9	21.7	12.873
12926	85.17	18.8	0	0	25.9	33.1	27.602
12927	87.87	13.8	0	0	25.6	18.7	21.632
12928	82.99	19.8	0	0	18.7	30.0	13.794
12929	84.32	18.0	0	0	6.9	16.4	27.126
13207	91.08	13.8	1	1	25.9	27.9	17.936
13208	86.78	12.0	1	0	24.6	0.0	21.614
13222	91.86	16.5	1	1	20.1	16.6	21.258
13223	90.64	12.5	1	1	26.5	23.0	20.278
13224	89.48	17.3	1	1	12.5	15.6	29.716

3. RESULTS AND CONCLUSION

Table 2 shows the results obtained from the fitting between sample preparation variables and the beta efficiency obtained from the $4\pi\beta-\gamma$ coincidence system.

Table 2 Parameters obtained in the multi-parametric fitting between beta efficiency and sample preparation variables.

Parameter	a_0 Intercept	a_1 Collodion Film thickness	a_2 Gold Deposit Side 1 (µg/cm^2)	a_3 Gold Deposit Side 2 (µg/cm^2)	a_4 Radioactive source mass (mg)	a_5 Evaporation Atmosphere (warm nitrogen jet)	a_6 Seeding Agent (with Cyastat)
Value	81.5	-0.096	0.147	0.018	0.064	2.86	3.12
Standard Deviation	1.9	0.097	0.046	0.035	0.048	0.81	0.90

As can be seen the most important parameters were: 1- Use of warm nitrogen jet as the evaporation atmosphere; 2- Application of seeding agent (Cyastat) and 3- Gold deposition on side one. The parameters associated with source mass and gold deposit on side 2 show uncertainties close to their values, indicating that they are irrelevant within the statistics of the experiment. It can also be concluded that covering only one side of the film with gold is enough for rendering the highest beta efficiency. The influence of the Collodion film thickness is zero within the statistical uncertainty, indicating negligible influence in the efficiency, for the thickness range of the present experiment (9,0 µg/cm^2 – 21,0 µg/cm^2).

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