

## A COINCIDENCE SYSTEM FOR RADIONUCLIDE STANDARDIZATION USING SURFACE BARRIER DETECTORS

Marina F. KOSKINAS and Mauro S. DIAS

*Instituto de Pesquisas Energéticas e Nucleares (IPEN), Comissão Nacional de Energia Nuclear – CNEN/SP, Caixa Postal 11049, CEP 05499, São Paulo, Brazil*

A system for the standardization of alpha–gamma or electron–X radionuclide emitters is described. The system consists of one or two surface barrier detectors for alpha or electron detection which are coupled to thin-window NaI(Tl) crystals suitable for low-energy X- or gamma-ray detection. The performance of the system has been verified by the standardization of  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{109}\text{Cd}$  solutions. The activity has been obtained using the extrapolation method applied to the  $4\pi$   $\alpha$ - $\gamma$  and  $2\pi$   $e$ -X coincidence techniques. The surface barrier detection efficiency was varied by placing absorbers over the radioactive source or by changing the source-to-detector distance. The results were compared to those obtained using conventional absolute systems based on gas-flow and pressurized  $4\pi$  proportional counters, or using radioactive solutions standardized in international comparisons sponsored by the Bureau International des Poids et Mesures, France. The expected and measured activities agreed within the experimental uncertainties, which were: 0.2% for  $^{241}\text{Am}$ , 0.7% for  $^{137}\text{Cs}$  and 0.6% for  $^{109}\text{Cd}$ .

### 1. Introduction

Gas-flow or pressurized proportional counters in  $4\pi$  geometry are commonly used for the absolute standardization of radionuclides, either measuring single rates or in coincidence with some other detector, usually a NaI(Tl) scintillator counter [1–3]. High accuracy can be achieved in many cases by using the extrapolation technique [4–6]. However, the sample preparation requires a substrate coated with a conducting material and sources with high activity cannot be standardized directly due to dead-time losses.

Liquid scintillator counters are widely used [7,8] and can measure higher decay rates in comparison to proportional counters. However, the sample preparation is destructive and the standardized source cannot be used to calibrate other relative counting systems, e.g. HPGe or surface barrier detectors with defined geometry. Moreover, liquid scintillators are by far more sensitive to gamma radiation and have a higher background in comparison to surface barrier detectors.

The present work describes an alternative absolute counting system where the sample preparation is simpler. High-activity sources can be measured, which may be used to calibrate relative counting systems.

The absolute system consists of surface barrier detectors operating in coincidence with thin-window NaI(Tl) detectors. The performance of the system has been verified by measuring three radionuclides of different decay-scheme characteristics, namely:  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{109}\text{Cd}$ . The results were compared to those obtained by conventional absolute systems or in international com-

parisons sponsored by the BIPM (Bureau International des Poids et Mesures, France).

### 2. Counting apparatus

A schematic view of the detection setup is shown in fig. 1. One or two surface barrier detectors can be used, either with conventional geometry (back-sided connector) or with transmission detectors. The setup allows the variation of the source–detector distance from 0.3 to 15 mm symmetrically with respect to the source. The NaI(Tl) detector holder has similar features and the system can also be used with scintillators alone for  $\gamma$ - $\gamma$  or X–X coincidence measurements [9].

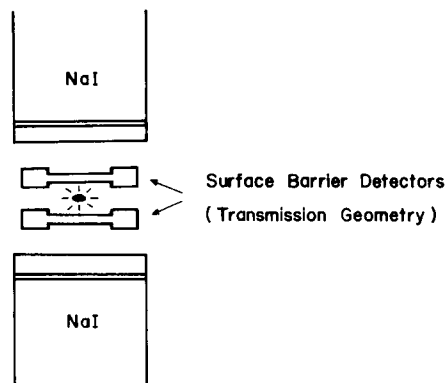


Fig. 1. Schematic view of the surface-barrier–NaI(Tl) coincidence system.

For alpha–gamma emitters a nearly  $4\pi$  geometry was achieved by employing two transmission surface barrier detectors with 200 mm<sup>2</sup> of active area and 1000  $\mu\text{m}$  depletion depth. Comparisons were made employing a single detector approaching a  $2\pi$  geometry.

For electron–X-ray coincidence measurements a single barrier detector was used in order to avoid the attenuation of the X-ray beam striking the scintillation counter.

The X- and gamma-rays were detected by a pair of NaI(Tl) crystals 1 mm thick and with a 0.025 mm aluminium window. The measured resolution of these crystals was around 44% at an energy of 5 keV.

The electronics consisted of two counter chains with 3  $\mu\text{s}$  fixed dead time and a coincidence with a resolution time of 1  $\mu\text{s}$ . The single counting rates were corrected for dead time and the coincidence rate was corrected by the Cox–Isham formula [10].

### 3. Sample preparation

The sources were prepared by dropping known aliquots of the radioactive solution on a 20  $\mu\text{g}/\text{cm}^2$  thick Collodion film. This film was previously coated with a 10  $\mu\text{g}/\text{cm}^2$  gold layer in order to turn the film conducting and make possible to measure the same sources in a conventional  $4\pi$  proportional counter. Naturally, other types of source holders can be used, for instance electrodeposited sources with metal backing suitable for measurements of alpha emitters using a single surface barrier detector approaching  $2\pi$  geometry.

A seeding agent (Cystat SM) was used to improve the deposit uniformity and the sources were dried by a warm nitrogen jet [11]. The picnometer technique was used for accurate source mass determination [12].

### 4. The coincidence equations

A set of equations has been developed in order to obtain the desintegration rate for each of the three standardized nuclides, namely <sup>241</sup>Am, <sup>137</sup>Cs and <sup>109</sup>Cd.

The radionuclide <sup>241</sup>Am decays by alpha particle emission followed by several gamma rays [13]. The alpha particles were detected by the surface barrier detector whereas the NaI(Tl) detected the 59 keV gamma ray in coincidence with the alpha particles.

The equations for the three alpha, gamma and coincidence counting channels,  $N_\alpha$ ,  $N_\gamma$  and  $N_c$ , are those commonly used in the coincidence method [5]. Since the electronic discriminator for the alpha counting channel was set above 59 keV, the equations are greatly simplified and the desintegration rate is given by

$$N_0 = N_\alpha N_\gamma / N_c.$$

In the case of <sup>137</sup>Cs the surface barrier detector was set to count only the K + L + ... conversion electrons [14]. The NaI(Tl) detector was set to count the K X-rays following the internal conversion process.

The <sup>137</sup>Cs desintegration rate is simply given by

$$N_0 = N_e N_X / N_c P_1,$$

where  $N_e$ ,  $N_X$  and  $N_c$  are the electron, X-ray and coincidence counting rates and

$$P_1 = a \frac{1}{1 + \alpha_t} \left[ \alpha_K + (\alpha_t - \alpha_K) \frac{\epsilon_{ec}(L, M, N)}{\epsilon_{ec}(K)} \right].$$

Two correction factors had to be considered in the observed counting rates. The first is the 1174 keV beta-ray contribution in the conversion-electron counting channel which was subtracted by means of a spectrum fitting procedure. The second is the Compton scattering produced by the 661 keV gamma ray in the NaI(Tl) scintillator at the region of the <sup>137</sup>Ba K X-rays. This was corrected by making measurements covering the NaI scintillator with a 1 mm thick Cu absorber.

As in the case of <sup>137</sup>Cs, the surface barrier detector was set to count the K + L + ... conversion electrons and the NaI detector was set to count the K X-rays following the internal conversion process [15].

The equations for the counting channels in the case of <sup>109</sup>Cd are slightly different as compared to <sup>137</sup>Cs because of the contribution of K X-rays coming from the electron capture process to the NaI scintillator count rate. The activity is given by

$$N_0 = \frac{N_e N_X (P_2 - P_3)}{N_c P_1 P_2},$$

where  $P_2 = P_3 + \alpha_K \omega_K / (1 + \alpha_t)$  and  $P_3 = P_K \omega_K$ . A possible dependence of  $N_0$  on the alpha efficiency parameter  $N_c/N_\gamma$  (or  $N_c/N_X$ ) has been verified by placing absorbers over the source mount or by changing the source-to-detector distance. The value of  $N_0$  was obtained by extrapolation of  $N_c/N_\gamma$  to unity.

### 5. Experimental results

Fig. 2 shows the behaviour of  $N_\alpha N_\gamma / N_c$  as a function of  $(1 - N_c/N_\gamma) / (N_c/N_\gamma)$  for <sup>241</sup>Am. The closed circles are experimental points obtained in a  $4\pi$  geometry (two silicon detectors), and the open circles were obtained in a  $2\pi$  geometry (one silicon detector). As expected, the slope of the curve is zero within the experimental uncertainty and the extrapolated  $N_0$ -value has been obtained with an uncertainty of 0.1%. No difference was observed between the results obtained with  $4\pi$  or  $2\pi$  geometry. Therefore a single silicon detector can be used without significant loss in the resulting accuracy.

The results of the activity value obtained within the proposed  $4\pi$   $\alpha$ - $\gamma$ -coincidence system are compared to

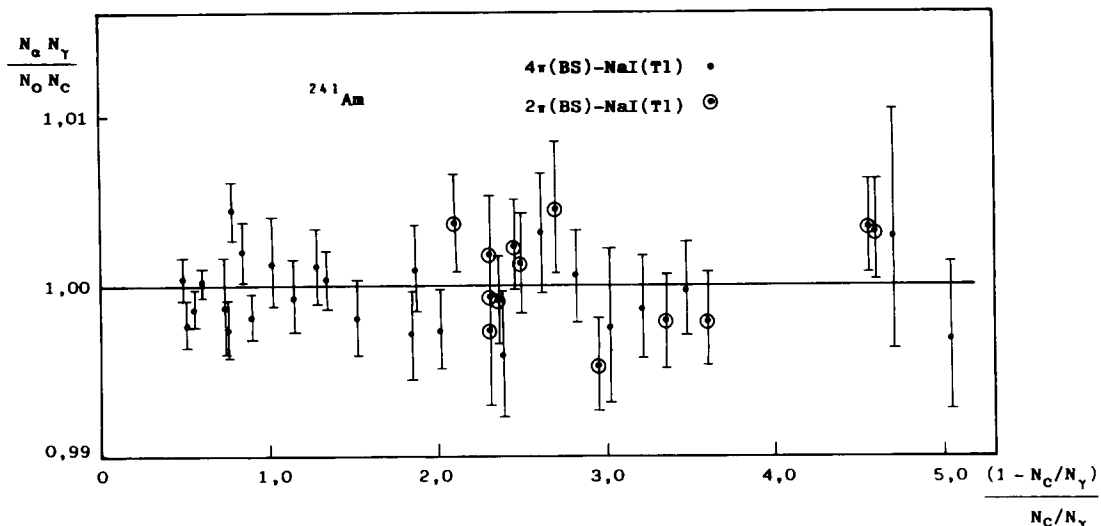


Fig. 2. Extrapolation curve for <sup>241</sup>Am desintegration-rate determination, normalized to unity for comparison between 4π and 2π geometries.

Table 1  
Results of the standardization of <sup>241</sup>Am

System	Specific activity [kBq/g]
4π (BS) - NaI(Tl) <sup>a)</sup>	150,3 ± 0,3
4π (BS) - NaI(Tl) <sup>b)</sup>	150,5 ± 0,3
4π (PC)	150,5 ± 0,3

<sup>a)</sup> Using external absorbers.

<sup>b)</sup> Changing source-detector distance.

Table 2  
Results of the desintegration rate of a <sup>137</sup>Cs solution

System	Specific activity [kBq/g]
2π BS - NaI(Tl)	604,1 ± 4,2
International comparison [16], 1982 <sup>a)</sup>	604,48 ± 2,17

<sup>a)</sup> Weighted average.

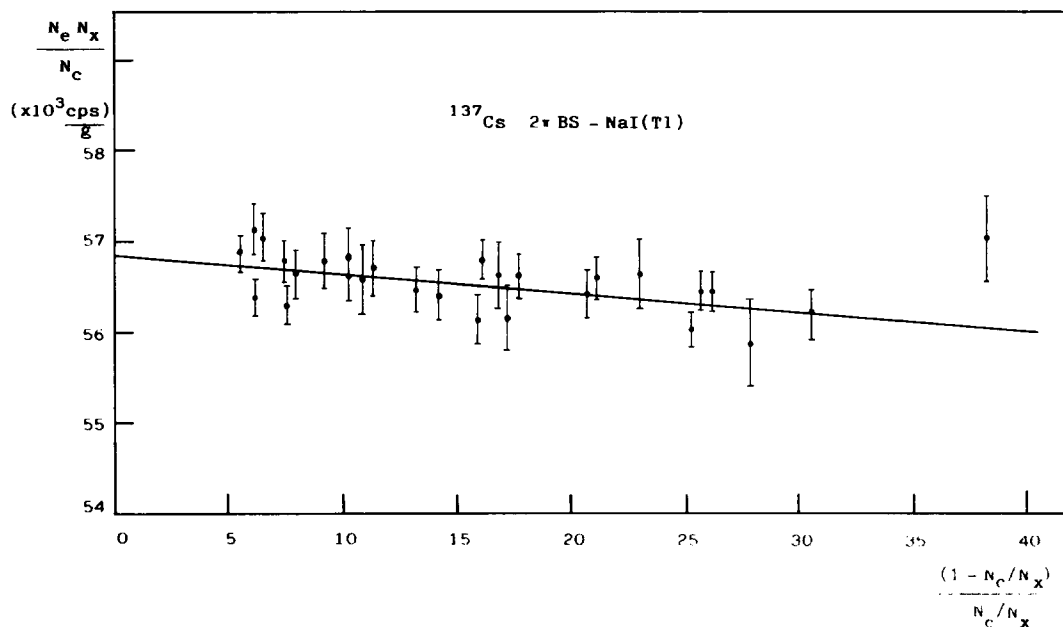


Fig. 3. Extrapolation curve for <sup>137</sup>Cs desintegration-rate determination.

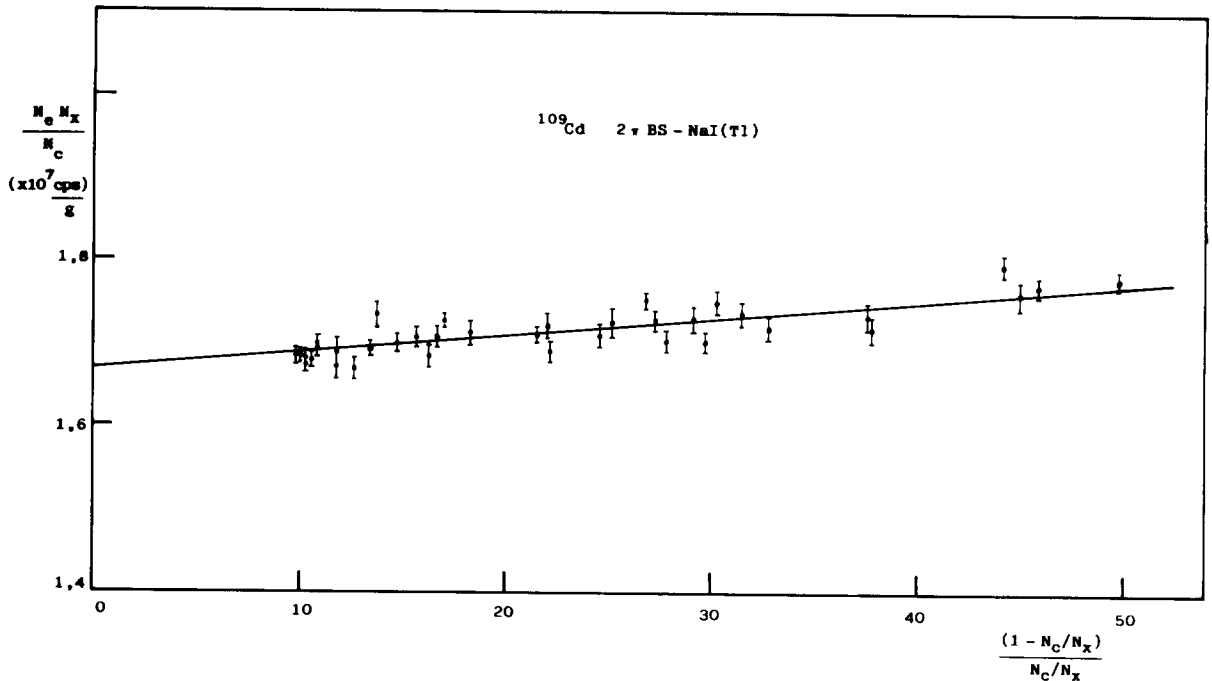


Fig. 4. Extrapolation curve for  $^{109}\text{Cd}$  desintegration-rate determination.

those obtained in a  $4\pi$  proportional counter in table 1. The results agree within the experimental uncertainty.

The extrapolation curve for  $^{137}\text{Cs}$  is shown in fig. 3. The activity results are shown in table 2 in comparison with those obtained in the 1982 international comparison of this radionuclide [16]. The results are in good agreement within the estimated experimental uncertainty.

The extrapolation curve for  $^{109}\text{Cd}$  is shown in fig. 4. The small slope observed was attributed to a change in the ratio  $\epsilon_{\text{ec}}(\text{L, M, N})/\epsilon_{\text{ec}}(\text{K})$  as the detector goes further from the source. The activity results are shown in table 3 in comparison with the one obtained in a  $4\pi$  pressurized proportional counter [17] and with the weighted average from the 1986 international comparison

of this radionuclide [18]. There is a good overall agreement. Because in this case the results for the proposed system are dependent on the decay scheme parameters, slightly different values are obtained using data from different references [15,19].

From the results of  $N_0 P_1 P_2 / (P_2 - P_3)$  for  $^{109}\text{Cd}$  obtained with the proposed system and the results of  $N_0 P_1$  obtained with the  $4\pi$  pressurized proportional counter, it was possible to determine the decay parameter  $P_2 / (P_2 - P_3)$ . This parameter resulted in  $2.8883 \pm 0.016$ , in good agreement with Kawada's value [19]  $2.8859 \pm 0.009$ .

## 6. Conclusions

The activity results obtained with the proposed  $4\pi$   $\alpha$ - $\gamma$  surface barrier coincidence system were in good agreement with those obtained with conventional absolute systems or in international comparisons for the three measured radionuclides  $^{241}\text{Am}$ ,  $^{137}\text{Cs}$  and  $^{109}\text{Cd}$ .

The accuracy achieved with a single silicon detector approaching  $2\pi$  geometry is similar to the one using two detectors in nearly  $4\pi$  geometry, provided the same statistical uncertainty is achieved at a longer measuring time.

In some cases, e.g.  $^{241}\text{Am}$ , the activity result does not depend on scheme parameters and therefore it can be obtained with high accuracy.

Table 3  
Results of the desintegration rate of a  $^{109}\text{Cd}$  solution

System	Specific activity [kBq/g]
$2\pi$ BS - NaI(Tl) <sup>a)</sup>	$5935 \pm 53$
$2\pi$ BS - NaI(Tl) <sup>b)</sup>	$5987 \pm 33$
$4\pi$ PPC e <sup>-</sup>	$6000 \pm 12$
International comparison [18] <sup>c)</sup>	$5992 \pm 6$

<sup>a)</sup> Using decay scheme parameter from ref. [15].

<sup>b)</sup> Using Kawada's value [19].

<sup>c)</sup> Weighted average.

The proposed system does not require conducting films for source mounts, simplifying appreciably the sample preparation. High-activity sources can be measured by placing the silicon detector further away from the source.

Because the extrapolation method by changing the source–detector distance is quite simple, standard sources of selected metastable nuclides can be prepared quickly and the attained accuracy is usually satisfactory for the calibration of secondary activity measurement systems.

### Acknowledgements

The authors are grateful to Miss E. Pocobi for the careful sample preparation and to Mr. C.A. Silva and Mr. R.R. Machado for the help in the data analysis.

### References

- [1] A.P. Baerg, Nucl. Instr. and Meth. 112 (1973) 95.
- [2] M.E.C. Troughton, Int. J. Appl. Radiat. Isot. 28 (1973) 773.
- [3] A.P. Baerg, Metrologia 3 (1967) 105.
- [4] P.J. Campion, Int. J. Appl. Radiat. Isot. 4 (1959) 232.
- [5] A.P. Baerg, Metrologia 2 (1966) 23.
- [6] R.H. Martin and J.G.V. Taylor, Appl. Radiat. Isot. 38(10) (1987) 781.
- [7] R. Vaninbroux, Nucl. Instr. and Meth. 112 (1973) 111.
- [8] R. Vatin, Monographie BIPM-3 (1980).
- [9] M.S. Dias and M.F. Koskinas, to be published.
- [10] D.R. Cox and V. Isham, Proc. R. Soc. London A356 (1977) 149.
- [11] H.A. Wyllie, E.P. Johnson and G.C. Lowenthal, Int. J. Appl. Radiat. Isot. 21 (1970) 497.
- [12] P.J. Campion, Monographie BIPM-1 (1975).
- [13] M.J. Martin and P.H. Blichert-Toft, Nucl. Data. Tables A8 (1970) 1.
- [14] P. Christmas and P. Cross, Metrologia 14 (1978) 157.
- [15] F. Lagoutine, N. Coursol and J. Legrand, Table des Radionuclides, CEA-LMRI-(1982).
- [16] A. Rytz, BIPM-82/14 (1983).
- [17] M.S. Dias and M.F. Koskinas, IX Reunião de Trab. Sobre Física Nuclear, Brasil (Aug./Sept. 1986).
- [18] G. Ratel, CCEMRI(II)/87-7 BIPM (1987).
- [19] Y. Kawada, ETL-730 (1972) 66.