PERGAMON

Applied Radiation and Isotopes 56 (2002) 301–305

Applied Radiation and Isotopes

www.elsevier.com/locate/apradiso

Radioactivity standardization in South Africa

B.R.S. Simpson*

Radioactivity Standards Laboratory, CSIR–NML, 15 Lower Hope Road, Rosebank 7700, Cape Town, South Africa

Accepted 31 July 2001

Abstract

South Africa's national radioactivity measurement standard is maintained at a satellite laboratory in Cape Town by the National Metrology Laboratory (NML) of the Council for Scientific and Industrial Research. Standardizations are undertaken by a number of direct methods utilizing liquid scintillation counting (LSC). The successful application of LSC to the $4\pi\beta-\gamma$ coincidence method is reviewed. The activity unit is maintained through radionuclide specific calibration factors relating to a pressurized re-entrant well type ionization chamber. A comparison is made between normalized factors given by the manufacturer and deduced factors obtained by a method used to transfer calibration factors from the International Reference System of the International Bureau of Weights and Measures based on the NML's own absolute standardizations.

Keywords: South Africa; Radioactivity measurement standard; Liquid scintillation coincidence counting; Ionization chamber radionuclide factors

1. Introduction

The realization of radioactivity measurement standards in South Africa has recently been re-established by the Council for Scientific and Industrial Research (CSIR), thereby fulfilling its statutory responsibility to maintain the national radioactivity measurement standard. The facility is operated as a satellite laboratory in Cape Town by the CSIR's Pretoria-based National Metrology Laboratory (NML). This paper reviews the development of the radioactivity programme.

Standardization of radioactivity in South Africa has a long history. It was originally established at the CSIR's National Physical Research Laboratory in the early 1950s (McMurray, 1960). The radioactivity standards laboratory existed in various guises at the CSIR Pretoria campus until, in 1981, it was transferred to the National Accelerator Centre (NAC) near Cape Town. From 1989 onwards, the laboratory maintained the national standard in a 'de facto' capacity after the management of the NAC was taken over from the CSIR by a newly created government body. A break in continuity occurred from mid 1999 to the end of 2000 when the NAC closed down the laboratory. This prompted the CSIR to purchase the equipment from the NAC and re-establish South Africa's capability in radionuclide metrology.

Right from the outset, an interest was taken in liquid scintillation counting (LSC). In fact, the application of internal LSC to the absolute standardization of aqueous solutions of β-emitters was developed at the CSIR laboratory (Steyn, 1956; Steyn and Haasbroek, 1958). The initial success achieved encouraged the laboratory to specialize in the utilization of LSC techniques for the accurate measurement of radioactivity. This tradition continues at the new facility.

2. Standardization of γ-emitting radionuclides

Since γ-emitting radionuclides predominate, most standardizations undertaken by the laboratory have utilized the $4\pi\beta-\gamma$ coincidence extrapolation method (Baerg, 1973). The locally built counting system is based on internal LSC to detect the electrons from β-emitters

*Tel.: +27-21-686-2759; fax: +27-21-686-6116.
E-mail address: bsimpson@csir.co.za (B.R.S. Simpson).

0969-8043/02/$ - see front matter
PII: S0969-8043(01)00205-6

as well as from those nuclides decaying by electron capture (EC). The source material is dissolved in 12 ml of liquid scintillator, the counting vial being viewed by two RCA 8850 high-gain phototubes connected in coincidence to eliminate random electronic noise. The γ-rays are detected with a $75 \times 75\,mm^2$ NaI(Tl) crystal. The signal processing electronics allows for the setting of multiple thresholds in the β pulse-height spectrum that enables up to 15 datum points of varying efficiency to be simultaneously collected for each source (Simpson and Meyer, 1988). A computer programme operating in the Windows® environment is used to control the counting system and record the data (Simpson and van Oordt, 1997).

The scintillator has a significant detection efficiency for γ radiation, leading to γ–γ coincidences from γ-rays being Compton scattered out of the scintillator and being observed by the γ detector. To avoid these, a γ pulse height window is usually set to select only the photopeak events from the highest energy γ-ray. This criterion helps simplify theoretical detection efficiency analyses undertaken to confirm that the method is justifiable for the radionuclide of interest. Fig. 1 shows an example of the effect of γ–γ coincidences (Simpson and Meyer, 1993), achieved by including the Compton portion of the γ spectrum in the γ window. Theoretical analysis (Steyn and Haasbroek, 1958; Simpson and Meyer, 1993) explains the observed change in slope and also verifies that the source activity is extracted independently of the γ window setting when extrapolating to an efficiency equal to unity.

Contrary to common practice, the philosophy has been to rather not introduce an artificial fixed dead time into the system, thereby keeping the dead time corrections as small as possible. The intrinsic dead time is essentially given by the amplifier pulse width, which is of the order of 1 μs. The actual dead times are measured by a variation of the paired-source method (Simpson, 1991). Together with the coincidence resolving times involved (typically 0.47 μs), the rate-dependent corrections are determined by a simple formula derived by Bryant (1963) that provides accurate results up to 30,000 counts per second.

Table 1 gives the results of standardized samples submitted to the International Reference System (SIR), a comparison programme operated by the International Bureau of Weights and Measures (BIPM), (Rytz, 1983). Comparison against the SIR mean values, which are almost entirely made up of non-LSC techniques, shows good agreement throughout. Comparable results were also obtained in recent international comparisons (Table 2).

3. Standardization of non-γ-emitting radionuclides

In recent years, a triple-phototube LSC system has been assembled for the activity measurement of pure β-emitters and pure EC nuclides. A variation of the triple-to-double coincidence ratio technique has been

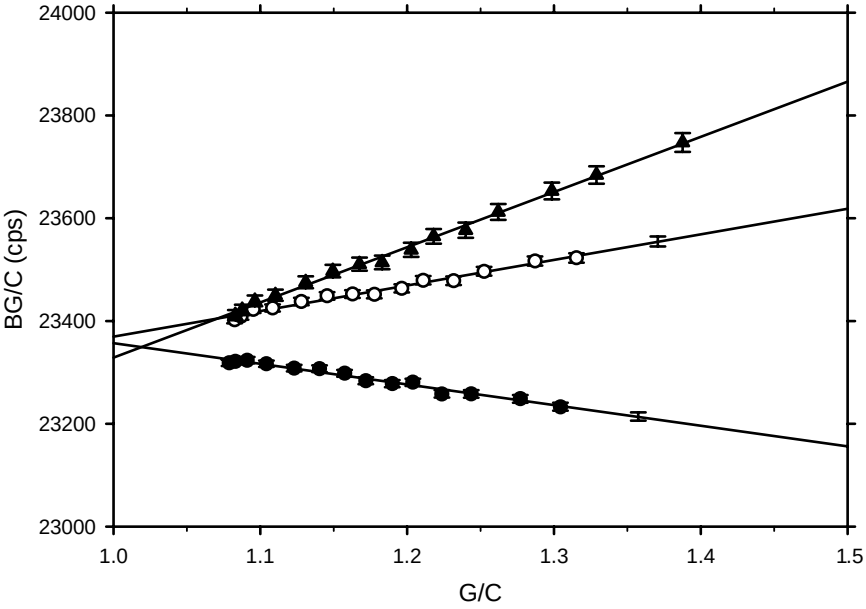

Fig. 1. A series of 4π count rates, B, from a ^{60}Co liquid scintillation source expressed as a function of the inverse of the β efficiency, G/C, where G is the γ-channel rate and C the coincidence rate. The lower set (closed circles) corresponds to a γ-ray threshold placed just above the noise, the middle set (open circles) had the threshold located midway in the Compton portion of the spectrum and the upper set (triangles) had the threshold set just below the 1.333 MeV photopeak. The solid lines are linear fits to the data. The variation in the extrapolated activity N from the three sets is consistent with counting statistics.

Table 1
Standardized samples submitted to the BIPM for comparison in the SIR, showing the measured "equivalent activity" A_e. Also shown are the average values computed from the *most recent* results of national laboratories participating in the SIR

Radionuclide	Decay type	Year	Activity A_e (kBq)	SIR mean A_e (unweighted)	SIR mean A_e (weighted)
^{22}Na	β^+	1984	7527±15	7540±15	7536±13
^{57}Co	EC	1985	170700±450	167842±1086	168832±832
^{60}Co	β^-	1992	7066±10	7064±4	7065±3
^{67}Ga	EC	1986	116413±292	116215±453	116073±250
^{75}Se	EC	1990	41998±593	43045±301	43114±188
^{109}Cd	EC	1982	8171000±59000	8114750±30725	8120054±19982
^{131}I	β^-	1980	40429±204	40349±25	40387±26
^{139}Ce	EC	1999	133100±850	132248±793	132891±444
^{201}Tl	EC	1991	312660±1980	311037±2577	312729±2486

Table 2
Results of the South African 4π(LS)$\beta-\gamma$ activity measurements from recent BIPM international comparisons. Except for ^{204}Tl, the mean value comprises mostly non-LSC techniques

Radionuclide	Decay type	Year	Activity (kBq/g)	Comparison mean (kBq/g)	References
^{133}Ba	EC	1984	1162.8±3.7	1160.8±4.2	Ratel (1992)
^{109}Cd	EC	1986	6026±41	5992±7	Ratel (1994)
^{125}I	EC	1988	1447.6±7.6	1427.6±4.4	Ratel (1995)
^{75}Se	EC	1992	1231.8±3.5	1253±3	Simpson and Meyer (1995)
^{204}Tl	β^-, EC	1994[a]	55.40±0.33	55.30±0.18	Simpson and Meyer (1996)

[a] Trial comparison (Ratel, 1999).

developed (Simpson and Meyer, 1994) and good results have been obtained in recent comparisons of ^{3}H (Makepeace et al., 1998), ^{63}Ni (Cassette et al., 1998) and ^{204}Tl (Ratel, 1999). The CIEMAT/NIST method (Coursey et al., 1985) is also sometimes applied and a variation of this method based on ^{54}Mn (Simpson and Meyer, 1998) has been used to accurately measure ^{55}Fe.

The activity of the low energy emitter ^{125}I is measured by photon–photon coincidence counting with a dual NaI(Tl) detection system (Simpson and Meyer, 1989; Ratel, 1995).

4. Specific precautions taken in applying the LSC methods

A significant advantage of using the liquid scintillation medium to prepare counting sources is the absence of self absorption problems (Steyn and Haasbroek, 1958). This advantage is somewhat offset by two effects associated with LSC, namely source instability and afterpulsing. These phenomena are always monitored and accounted for when necessary.

4.1. Preparation of reliable counting sources

For the triple-phototube system, counting sources are prepared in commercial glass vials. Custom made flat-faced cylindrical sample containers (Steyn, 1967) are preferred for use with the two-phototube system. These counting cells are directly coupled to the faces of the phototubes, thus providing increased efficiency. An effect that can lead to unreliable measurements is the adsorption of the radioactive material to the walls of the counting vials. At the NML, this is overcome by the introduction into the scintillation cocktail of a sufficient amount of appropriate carrier and/or acid solution. In addition, the vial is sometimes first saturated with carrier so that the stable form of the nuclide is preferentially adsorbed. Although these remedies lead to a reduction in counting efficiency due to chemical quenching, this does not impact since the resulting efficiencies are accurately determined. In the $4\pi\beta-\gamma$ method, any *residual* adsorption ($\leqslant 1\%$) in the counting vial does not compromise the measured activity because the loss in the 4π rate is inherently accounted for by a lowering in the measured efficiency. However, a compensating correction must be applied if any residual adsorption

Table 3
Radionuclide factors obtained at the NML from standardized samples. These are compared against those supplied with the ionization chamber (NPL) and those derived from the SIR "equivalent activity" values, A_e[a]

Nuclide[b]	Mean A_e	NML factor	NPL factor[c]	SIR deduced factor	Difference from NPL value [%]
^{24}Na	4963±4	—	8708±1%	8693	−0.17
^{88}Y	6907±13	—	6258±1%	6259	+0.02
^{60}Co	7061±3	6118±0.3%	6118±0.5%	6124	+0.10
^{22}Na	7529±8	5789±0.5%	5783±1%	5746	−0.64
^{46}Sc	8323±6	—	5173±1%	5202	+0.56
^{134}Cs	10097±10	—	4294±2%	4296	+0.05
^{154}Eu	13778±33	—	3195±2%	3160	−1.1
^{59}Fe	14661±17	—	2945±1%	2973	+0.95
^{152}Eu	14921±19	—	3074±2%	2922	−4.9
^{58}Co	16275±24	—	2667±2%	2682	+0.56
^{192}Ir	19011±52	—	2336±2%	2303	−1.4
^{54}Mn	19223±11	—	2252±1%	2278	+1.2
^{137}Cs	27563±38	1575±1.5%	1590±2%	1602	+0.75
^{65}Zn	29608±80	—	1449±2%	1495	+3.2
^{85}Sr	30023±29	—	1454±2%	1475	+1.4
^{131}I	40373±36	1107±1%	1107±1%	1108	+0.09
^{111}In	42812±112	—	1144±2%	1048	−8.4
^{75}Se	42815±164	1106±2%	1089±5%	1048	−3.8
^{133}Ba	43681±191	—	1180±2%	1028	−12.9
^{139}Ce	132280±834	471±2%	471±2.5%	370	−21.0

[a] The mean A_e values were based on *all* submissions to the SIR.
[b] Highlighted are NML samples sent to the SIR for comparison.
[c] Normalized to the NML's factor for ^{60}Co.

occurs when employing the methods for standardizing non-γ-emitters.

4.2. Afterpulsing

Although the use of two phototubes in coincidence considerably reduces afterpulsing, a correction of up to 1% may still be required especially if the β discrimination level is set below or within the single electron peak. At the NML afterpulsing is measured by a method based on a comparison of count rates corresponding to the intrinsic dead time τ_D and a considerably longer non-extending dead time, τ, that is introduced (Steyn and Botha, 1976). This is achieved by the use of a positive logic signal with the interval set from the start of a genuine pulse until reset time. The signal width chosen corresponds to $\tau \approx 94\,\mu s$, during which time most of the afterpulsing will have occurred (Smith, 1980). The probability, θ, for the production of one or more spurious pulses per genuine pulse is given by (Steyn and Botha, 1976)

$$\theta = \frac{q - n_0}{n_0} - (\tau - \tau_D)\frac{q}{T},$$

where T is the counting period, q is the total number of counts corresponding to τ_D and n_0 the number of counts associated with the longer dead time.

5. Maintaining the standardizations with an ionization chamber

The activity unit is maintained through radionuclide specific calibration factors relating to a re-entrant well type ionization chamber (IC), details of which can be found in Reher et al. (1998). Radionuclide factors for a number of nuclides, all standardized by the $4\pi(LS)\beta-\gamma$ extrapolation method, have been obtained from samples comprising 3.6 ml of solution in a 5 ml ampoule. Table 3 compares these factors with factors given by the IC manufacturer (provided by the National Physical Laboratory (NPL), Teddington, UK). The NPL values were scaled from those given for 3 ml solutions in 5 ml British Standard ampoules so as to be normalized to the NML's ^{60}Co value. Also shown are predictions obtained by a method used to deduce factors from the SIR (Reher et al., 1998), based on the four NML SIR-validated samples ^{75}Se, ^{137}Cs, ^{22}Na and ^{60}Co. It is evident that for lower energies the deduced factors become unreliable, probably due to different responses of the ICs at these energies.

6. Conclusions

It has been demonstrated through participation in the SIR and BIPM international comparisons, in which

mostly non-LSC techniques were employed, that there is no doubt as to the viability of applying LSC to the $4\pi\beta-\gamma$ extrapolation method. The IC radionuclide factors obtained from a limited number of these LSC standardizations are all in good agreement with normalized factors supplied by the manufacturer, giving confidence that the other factors are also applicable to the NML chamber. This notion is supported by factors independently deduced from a method based on transference from the SIR.

References

Baerg, A.P., 1973. The efficiency extrapolation method in coincidence counting. Nucl. Instrum. Methods 112, 143.

Bryant, J., 1963. Coincidence counting corrections for dead-time loss and accidental coincidences. Int. J. Appl. Radiat. Isot. 14, 143.

Cassette, P., Altzitzoglou, T., Broda, R., Collé, R., Dryák, P., de Felice, P., Günther, E., Los Arcos, J.M., Ratel, G., Simpson, B., Verrezen, F., 1998. Comparison of activity concentration measurement of ^{63}Ni and ^{55}Fe in the framework of the EUROMET 297 project. Appl. Radiat. Isot. 49, 1403.

Coursey, B.M., Grau Malonda, A., García-Toraño, E., Los Arcos, J.M., 1985. The standardization of pure-beta-particle-emitting radionuclides. Trans. Am. Nucl. Soc. 50, 13.

Makepeace, J., Altzitzoglou, T., Cassette, P., Dryák, P., Günther, E., Verrezen, F., Broda, R., Simpson, B., Unterweger, M., 1998. International comparison of measurements of the specific activity of tritiated water. Appl. Radiat. Isot. 49, 1411.

McMurray, W.R., 1960. The national physical research laboratory's radioactivity division. Atomics and Energy 5(10).

Ratel, G., 1992. Activity concentration of a solution of ^{133}Ba: results of an international comparison. Nucl. Instrum. Methods Phys. Res. A 316, 318.

Ratel, G., 1994. International comparison of activity measurements of a solution of ^{109}Cd. Nucl. Instrum. Methods Phys. Res. A 345, 289.

Ratel, G., 1995. Activity concentration of a solution of ^{125}I: results of an international comparison. Nucl. Instrum. Methods Phys. Res. A 366, 183.

Ratel, G., 1999. Trial comparison of activity measurements of a solution of ^{204}Tl. Rapport BIPM-97/3. Bureau International des Poids et Mesures, Sèvres, France.

Reher, D.F.G., Woods, M.J., Simpson, B.R.S., Ratel, G., 1998. Portability of the calibration of SIR of BIPM to other ionization chambers for radioactivity measurements. Appl. Radiat. Isot. 49, 1417.

Rytz, A., 1983. The international reference system for activity measurements of γ-ray emitting nuclides. Int. J. Appl. Radiat. Isot. 34, 1047.

Simpson, B.R.S., 1991. Deadtime measurement and associated statistical uncertainty by means of a two-detector paired-source method. Appl. Radiat. Isot. 42, 811.

Simpson, B.R.S., Meyer, B.R., 1988. A multiple-channel 2- and 3-fold coincidence counting system for radioactivity standardization. Nucl. Instrum. Methods Phys. Res. A 263, 436.

Simpson, B.R.S., Meyer, B.R., 1989. The half-life of ^{125}I. Appl. Radiat. Isot. 40, 819.

Simpson, B.R.S., Meyer, B.R., 1993. Standardization of ^{60}Co by the 4π(LS)$\beta-\gamma$ coincidence method. NAC Report NAC/93-05. National Accelerator Centre, Faure, South Africa.

Simpson, B.R.S., Meyer, B.R., 1994. Direct activity measurement of pure beta-emitting radionuclides by the TDCR efficiency calculation technique. Nucl. Instrum. Methods Phys. Res. A 339, 14.

Simpson, B.R.S., Meyer, B.R., 1995. ^{75}Se: activity measurements by 4π(LS)e, X$-\gamma$ coincidence counting and International comparison of results. NAC Report NAC/95-02. National Accelerator Centre, Faure, South Africa.

Simpson, B.R.S., Meyer, B.R., 1996. Activity measurement of ^{204}Tl by direct liquid scintillation methods. Nucl. Instrum. Methods Phys. Res. A 369, 340.

Simpson, B.R.S., Meyer, B.R., 1998. Activity measurement of ^{55}Fe by an efficiency calculation method. Appl. Radiat. Isot. 49, 1073.

Simpson, B.R.S., van Oordt, E.J., 1997. Data acquisition program for the NAC radioactivity standards laboratory. NAC Report NAC/97-03. National Accelerator Centre, Faure, South Africa.

Smith, D., 1980. The problem of afterpulses. In "The application of liquid-scintillation counting to radionuclide metrology" (Monographie BIPM-3, Bureau International des Poids et Mesures, Sèvres) 65.

Steyn, J., 1956. Absolute standardization of beta-emitting isotopes with a liquid scintillation counter. Proc. Phys. Soc. A 69, 865.

Steyn, J., 1967. Internal liquid scintillation counting applied to the absolute disintegration-rate measurement of electron capture nuclides. Proceedings of the Symposium on Standardization of Radionuclides. IAEA-Proc. Series SM-79, IAEA, Vienna, p. 35.

Steyn, J., Botha, S.M., 1976. International comparison of the radioactivity of a ^{139}Ce solution (March 1976): the effect of spurious pulses on the accuracy of the final result. NPRL Research Report FIS 102. Council for Scientific and Industrial Research, Pretoria, South Africa.

Steyn, J., Haasbroek, F.J., 1958. The application of internal liquid scintillation counting to a 4π beta–gamma coincidence method for the absolute standardisation of radioactive nuclides. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Vol. 21. Geneva, New York, United Nations, p. 95.