Available online at www.sciencedirect.com

Applied Radiation and Isotopes 64 (2006) 1510–1514

Applied Radiation and Isotopes

www.elsevier.com/locate/apradiso

The IFIN-HH triple coincidence liquid scintillation counter

A.C. Razdolescu[a,*], R. Broda[b], P. Cassette[c], B.R.S. Simpson[d], W.M. Van Wyngaardt[d]

[a]*National Institute of R&D for Physics and Nuclear Engineering "Horia Hulubei"- IFIN-HH, Atomistilor str. 407, POB MG-6, Bucharest, Romania*
[b]*Radioisotope Centre POLATOM, 05-400 Otwock-Świerk, Poland*
[c]*Laboratoire National Henri Becquerel- CEA/DIMRI, BP 52, F-91193, Gif sur Yvette, France*
[d]*Radioactivity Standards Laboratory, CSIR NML, 15 Lower Hope Road, Rosebank 7700, Cape Town, South Africa*

Abstract

The paper summarizes the IFIN-HH triple coincidence liquid scintillation counter used for the implementation of the TDCR method. The electronic unit was recently extended to record the three individual double coincidence ratios to take into account the differences in the quantum efficiencies of the three-photomultiplier tubes. Some details of the electronic system and the data processing are given.

The critical point of a TDCR counter is to adjust correctly the discriminator levels on the three channels under the single electron peak. The paper describes the method of adjustment based on the evolution of the dark counting rate versus the discriminator level. Also indicated is the influence of the discrimination level on the activity results as measured at IFIN-HH using a ^{3}H standard.

The performances of the IFIN-HH TDCR counter was checked against the measurement results of the TDCR counters of CSIR NML (South Africa), RC (Poland) and LNHB (France). A set of ready-to-measure ^{63}Ni sources in liquid scintillator, in sealed counting vials, was prepared and dispatched for measurement to all these laboratories. The paper describes designs of the TDCR counters used. An analysis and discussion of the measurement results is given.

© 2006 Elsevier Ltd. All rights reserved.

Keywords: TDCR method; Discrimination level; Comparison; ^{63}Ni sources

1. Introduction

The triple to double coincidence ratio (TDCR) method is an absolute activity measurement method by liquid scintillation counting (LSC) which was described in many previous papers (Vatin, 1991; Cassette and Vatin, 1992; Broda and Pochwalski, 1992; Meyer and Simpson, 1990). The TDCR method uses a detector with three photomultiplier tubes (PMTs), allowing the observation of double coincidences from two PMTs and triple coincidences (T) from three PMTs, as well as obtaining the logical sum of double coincidences (D) (Razdolescu and Cassette, 2004). The *TDCR* parameter, being a ratio of T to D, is a convenient indicator of the detection efficiency (or quenching) of the source.

Recently, the electronic unit in the IFIN-HH TDCR system was extended by additional scalars to record simultaneously a full set of coincidence count rates. Working parameters of the system were set to fulfill the requirements of the TDCR method. An important point was to ensure registration of all pulses resulting from the disintegration of the measured radionuclide, including single-electron pulses. The method of the correct adjustment of the discriminator levels is described below.

It was decided to validate the TDCR method used in the IFIN-HH system by an inter-laboratory comparison of measurement of activity of a ^{63}Ni solution. Laboratories of CSIR NML (South Africa), RC POLATOM (Poland) and LNHB (France) participated in this comparison. A set of ready-to-measure ^{63}Ni sources was dispatched to all participants. Results of measurement were collected and analyzed by using the same version of counting program. This procedure ensured the minimization of various factors influencing the results. The comparable results of the

---

[*] Corresponding author.
E-mail address: razdo@yahoo.com (A.C. Razdolescu).

0969-8043/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2006.02.070

extracted activity values validated the IFIN-HH TDCR system.

2. The IFIN-HH TDCR counting system

The IFIN-HH TDCR detection system contains three EMI 9804A PMTs placed at 120° angles from each other. The liquid scintillation source is placed in a plastic measurement chamber with the inner walls painted with a highly reflective paint. A high voltage of 1800 V for each PMT was provided by a CANBERRA 3105 high voltage power supply. The PMT outputs are connected to 612AM LeCroy® fast amplifiers. The coincidence module MAC3, constructed in LNHB (Bouchard and Cassette, 2000), accepts the signals from the three detection paths (A, B, C). An extending-type dead time signal with a minimum value of 40 μs is used. The NE4624 timer enabled live-time registration is to be applied. The count rates of three individual double coincidences (*AB*, *BC*, *AC*), a triple coincidence (*T*) and a logic sum of the three double coincidences (*D*) are registered simultaneously by two double CANBERRA 2071A, two double CANBERRA 2070 and one double CANBERRA 2071 scalars. Registration of the *AB*, *BC*, and *AC* counting rates enables one to take into account the differences in the quantum efficiencies of the three PMTs. Count rates are measured several times to obtain a single counting point. The counting efficiency in the IFIN-HH TDCR system can be reduced by using coaxial gray filters around the vial. An auxiliary MEASURE3 program is used for preparation of the output file.

3. Tuning the counting system

The critical point of the TDCR counting system is the adjustment of the discriminator levels on the three channels. The discriminator level must be set below the single electron peak and above the PMT noise. Measurements of the dark counting rate versus the discriminator level for each PMT were performed. The results are presented in Fig. 1. The discriminator level of at least 50 mV is necessary for proper operation of the MAC3. The working threshold of 55 mV for PMTs A and C and of 95 mV for B was selected. To check the correctness of the discriminator settings the background spectrum was measured by the multichannel analyzer gated by a counting signal.

Measurements of a ^{3}H standard source were performed with various discrimination thresholds. The counting efficiency was changed by using gray filters and the activity was calculated by the TDCRB-02 program (Cassette, personal communication). It was observed that the selection of the discrimination threshold influences the result of the measurements and the slope of a set of counting points (Fig. 2). The threshold level of $A = 55$ mV, $B = 95$ mV and $C = 55$ mV, where the slope is minimal, was taken for the next measurements.

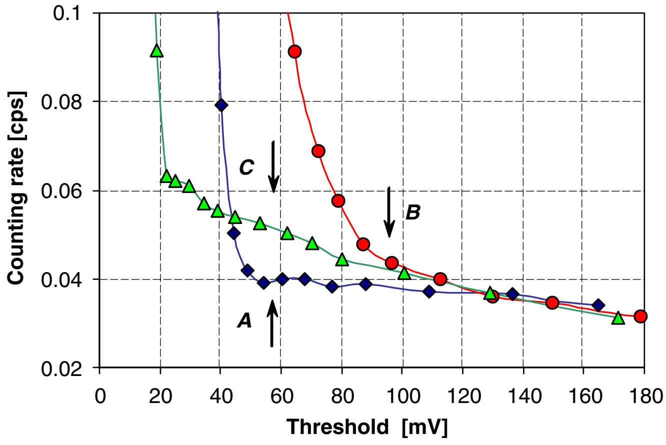

Fig. 1. Dark current counting rate versus the discriminator level (*A*, *B* and *C*) for three PMTs. Working thresholds are indicated by arrows.

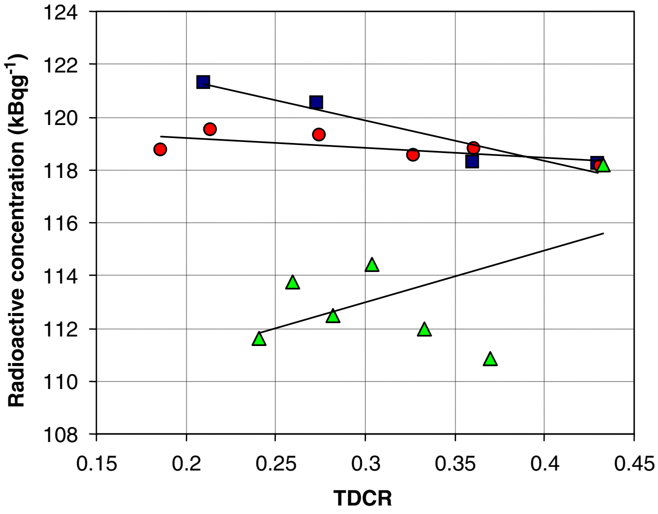

Fig. 2. Results of measurement of one ^{3}H source obtained with various discriminator levels (□—$A = B = C = 100$ mV; ○—$A = C = 55$ mV, $B = 95$ mV; Δ—$A = 68$ mV, $B = 55$ mV, $C = 65$ mV). Counting efficiency was changed by using gray filters.

The performance of the IFIN-HH TDCR system was checked against the results of the ^{63}Ni solution measurements at LNHB (France), RC POLATOM (Poland) and CSIR NML (South Africa) TDCR counters.

4. The LNHB TDCR system

The LNHB TDCR counter, previously described (Cassette and Vatin, 1992), is mainly composed of an optical chamber with three PMTs and a signal processing and interfacing unit. The BURLE 8850 type PMT, a 12-stage fast-linear-focused type with bialkali photocathode of 51 mm diameter and high gain at the first dynode, was used. PMT operated at 2200 V anodic voltage, which was found to be a good compromise between single electron pulse resolution and moderate dark counting rate. The anode of the three PMTs is connected to the MAC3 coincidence unit via fast preamplifiers and constant fraction discriminators. All information (i.e., non-coincident and coincident counting rates, live-time and real-time clock signals) is sent to CAMAC scalers. The CAMAC bus is connected to a PC via an IEEE488 interface.

The first dynode and focusing electrode can be adjusted in order to change the detection efficiency. All the high voltage power supplies are under the control of the PC, via the CAMAC bus.

The counting time is deduced from the number of pulses recorded in a live-time clock. Two independent clocks are used by the system: a 10 kHz quartz clock and a GPS-synchronized rubidium oscillator. The rubidium clock allows the traceability of the measurement to time units.

5. The RC POLATOM TDCR system

The RC POLATOM TDCR system (Chyliński et al., 2003) is very similar to that of the LNHB. It consists of a detection head with three PMT type RCA 8850 mounted horizontally, uniformly, at 120° angles around the liquid scintillation standard vial. The inner walls of the measurement chamber were coated with a white diffusing paint.

The high voltage 800 V at the first dynode and 2800 V for the anode of the three PMTs is applied. The coincidence module MAC3 is used. The minimal dead time of 63 μs is applied. A discrimination level below the single-electron peak pulse is set. The counting efficiency is modified by reducing the photomultiplier focusing electrode potential.

6. The CSIR NML TDCR system

The apparatus comprised two RCA 8850 PMTs (operated at + 1950 V) and one BURLE 8850 PMT (+ 2200 V). Two of the tubes were mounted at 180° and the third perpendicular to their axis in the same plane (Meyer and Simpson, 1990). The threshold of each channel was set in the valley below the single electron peak. Analogue signals were used and slow logic pulses were fed into a specially built TDCR module to provide three singles counts, three double-coincident counts, one logic sum of double-coincident count and one triple-coincident count. The signals were sent to a CAMAC data acquisition system interfaced to a PC. The non-extending dead time was used. The PMTs were efficiency matched, the three double rates agreeing to within 0.4% or better. The contribution from after-pulsing was monitored and recorded simultaneously with the measured coincidence rates (Simpson and Meyer, 1994; Simpson, 2002). The single counts were used only for rate-dependent corrections ($\tau_D = 1.0\,\mu s$, $\tau_R = 0.47\,\mu s$). The size of the double coincidence afterpulse correction was 0.13%.

7. Source preparation

A set of 16 ready-to-measure sources of the ^{63}Ni solution were prepared at LNHB. Stability of sources was checked over the course of one month. The sources were used for checking the performance of the IFIN-HH TDCR counting system by comparative measurements in LNHB, RC POLATOM and CSIR NML. Radioactive solution of 10 μg/g of $NiCl_2$ in HCl 0.1 M was mixed with 10 ml of Ultima Gold scintillator and closed in the sealed, standard, low potassium, 20 ml glass vials. The radioactive concentration of the ^{63}Ni solution was about 33 kBq/g at the reference time. The mass of radioactive ^{63}Ni solution in measured sources is given in Tables 1 and 2.

8. Results of ^{63}Ni measurements

All sources were measured at the LNHB by using the TDCR method. A set of 12 vials were dispatched for comparative measurement to IFIN-HH (source Nos. 1–4), to CSIR NML (sources Nos. 5–8) and to RC-POLATOM (sources Nos. 9–12). Measurements of each source were performed several times at the highest counting efficiency of the TDCR system only. Results were corrected for decay and background. In case of the CSIR NML TDCR system the count-rates were corrected for after-pulsing, dead time and coincidence resolving time.

The measurement data from all four laboratories were recalculated for the same reference time and analyzed using the TDCR09 program developed at LNHB, and which was based on the earlier version of a program by Broda et al. (2000), to eliminate possible discrepancies from different

Table 1
The comparative data of measurement of 3 ^{63}Ni sources at four TDCR systems, recalculated at the reference time using the same TDCR09 analysis program (T—triple coincidence, cps, BkgT—background for triple coincidence channel, cps, D—logical sum of double coincidences, cps, BkgD—background for double coincidences channel, cps, A_0—radioactive concentration (kBq g^{-1})

Source	Mass(mg)	Laboratory	TDCR	*T*(cps)	*BkgT*(cps)	*D*(cps)	*BkgD*(cps)	ε_D	A_o(kBq g^{-1})
1	90.671	LNHB	0.789	1879	5.6	2382	7.2	0.791	33.20
		IFIN	0.780	1824	2.3	2339	8.3	0.774	33.32
5	97.514	LNHB	0.793	2038	5.6	2570	7.2	0.794	33.20
		CSIR	0.780	1969	1.0	2523	3.7	0.772	33.50
9	96. 265	LNHB	0.789	1997	5.6	2532	7.2	0.791	33.27
		RC	0.782	1930	0.7	2469	3.8	0.772	33.21

Table 2
Radioactive concentration (A_o) of a set of 16 ^{63}Ni sources measured in four laboratories and calculated by the same TDCR09 program in each case

Source	Mass (mg)	IFIN-HH		CSIR NML		RC POLATOM		LNHB	
		A_o (kBq g^{-1})	u_C (kBq g^{-1})	A_o (kBq g^{-1})	u_C (kBq g^{-1})	A_o (kBq g^{-1})	u_C (kBq g^{-1})	A_o (kBq g^{-1})	u_C (kBq g^{-1})
1	90.671	33.32	0.1					33.20	0.05
2	119.558	33.24	0.1					33.16	0.05
3	106.418	33.26	0.1					33.33	0.05
4	99.180	33.14	0.1					33.21	0.05
5	97.514			33.50	0.12			33.20	0.05
6	93.784			33.58	0.12			33.23	0.05
7	97.426			33.54	0.12			33.22	0.05
8	102.534			33.13	0.12			33.13	0.05
9	96.265					33.21	0.09	33.27	0.05
10	96.019					33.11	0.09	33.27	0.05
11	104.508					33.20	0.09	33.24	0.05
12	102.184					33.28	0.09	33.25	0.05
13	98.347							33.23	0.05
14	98.159							33.22	0.05
15	93.319							33.27	0.05
16	92.025							33.26	0.05

Combined uncertainty (u_C) at 1 σ level.

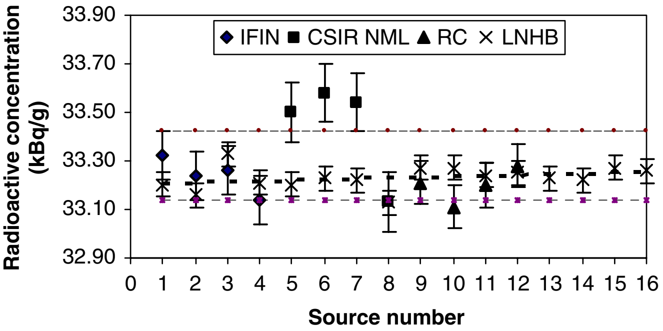

Fig. 3. Results of measurement of a set of 16 ^{63}Ni sources obtained in four laboratories. The same TDCR09 counting program was used in each case. Mean value of radioactive concentration of $33.28 \pm 0.14\,\mathrm{kBq\,g^{-1}}$ was found. Uncertainty limits of ±0.4% are shown.

analysis codes. The comparative data of all TDCR systems in case of the three selected ^{63}Ni sources are collected in Table 1. The triple-to-double coincidence ratio (*TDCR*), count rates of T and D channels, background in these two channels (*BkgT*, *BkgD*) as well as the counting efficiency in D channel (ε_D) are given. Results of measurement of radioactive concentration (A_o) of all ^{63}Ni sources from the set, performed at the four laboratories and computed by the same TDCR09 program, are presented in Table 2 and Fig. 3. Combined uncertainties (u_C) at 1 σ level are given.

The mean radioactive concentration for all sources is $A_o = 33.28\,\mathrm{kBq\,g^{-1}}$ with an uncertainty of $\pm 0.14\,\mathrm{kBq\,g^{-1}}$ (±0.4%). The difference between the highest and lowest measurement results obtained is 1%, which is acceptable. Equivalent results were obtained by using the TDCRB-02 version of the program.

9. Conclusions

The IFIN-HH TDCR system was successfully extended and tuned to fulfill the requirements of the TDCR measurement method. The procedure used for adjustment of the discriminator level under the single electron peak, based on the dark counting rates measurement, assured satisfactory working condition of the TDCR system.

The IFIN-HH TDCR system has been favorably validated by the inter-laboratory comparison of activity of the ^{63}Ni solution. The procedure adopted, where a set of ready-to-measure sources and the same program for analyzing of the results was used, ensured that only the operating conditions of all the TDCR systems could influence the results. Results of measurement of a set of 16 sources at four laboratories (IFIN-HH, CSIR NML, LNHB, RC POLATOM) were consistent within the given uncertainties. The spread of results was 1%.

Results obtained from the TDCR09 program can also be used for comparing against other locally developed versions of the analysis program used for the TDCR method.

References

Bouchard, J., Cassette, P., 2000. MAC3: an electronic module for the processing of pulses delivered by a three photomultiplier liquid scintillation counting system. Appl. Radiat. Isot. 52, 669–672.

Broda, R., Pochwalski, K., 1992. The enhanced triple to double coincidence ratio (ETDCR) method for standardization of radionuclides by liquid scintillation counting. Nucl. Instrum. Methods A312, 85–89.

Broda, R., et al., 2000. A simple computing program for application of the TDCR method to standardization of pure-beta emitters. Appl. Radiat. Isot. 52, 673–678.

Cassette, P., Vatin, R., 1992. Experimental evaluation of TDCR models for the 3 PM liquid scintillation counter. Nucl. Instrum. Methods A312, 95–99.

Chyliński, A., Broda, R., Radoszewski, T., 2003. The national standard unit of radionuclide activity and the related standards in Poland. Nukleonika 48 (1), 51–55.

Meyer, B.R., Simpson, B.R.S., 1990. A direct method for ^{55}Fe activity measurement. Appl. Radiat. Isot. 41, 375–379.

Razdolescu, A.C., Cassette, P., 2004. Standardization of tritiated water and ^{204}Tl by TDCR liquid scintillation counting. Appl. Radiat. Isot. 60, 493–497.

Simpson, B.R.S., 2002. Radioactivity standardization in South Africa. Appl. Radiat. Isot. 56, 301–305.

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 A339, 14–20.

Vatin, R., 1991. Mesure absolue de l'activite des radionucleides emetteurs beta purs par scintillateur liquide a l'aide d'un detecteur a 3 photomultiplicaleurs. CEA/DAMRI, Bulletin du BNM, 85.