

# Construction and implementation of the NIST triple-to-double coincidence ratio (TDCR) spectrometer

B.E. Zimmerman\*, R. Collé, J.T. Cessna

*Ionizing Radiation Division, Physics Laboratory, National Institute of Standards and Technology, Gaithersburg, MD 20899-8462, USA*

## Abstract

A description of the triple-to-double coincidence ratio (TDCR) spectrometer recently constructed at NIST and results of tests to assess its operating characteristics are presented. Measured activities for previously calibrated solutions of  $^3\text{H}$  (tritiated water) and  $^{63}\text{Ni}$  agreed with certified activity values to within 0.04% and 0.2%, respectively. Agreement between measurements of solutions containing  $^{90}\text{Y}$  using the TDCR and the CIEMAT/NIST efficiency tracing method was achieved to within 0.7% and within the respective experimental uncertainties. A difference of 5.3% was observed between TDCR measurements and the certified massic activity value for a calibrated solution containing  $^{54}\text{Mn}$ , but because of the large uncertainty on the TDCR result (3.6% relative standard uncertainty), the values agree to within the experimental uncertainties. Agreement to within experimental uncertainties (2%) for  $^{103}\text{Pd}$  was achieved with the TDCR and CIEMAT/NIST methods, but not between TDCR and isothermal calorimetric measurements. TDCR results for a  $^{204}\text{Tl}$  solution were in excellent agreement (maximum difference of 0.2% with any other method, with combined standard uncertainty of 0.45% on TDCR result) with results obtained using three other methods.

Published by Elsevier Ltd.

**Keywords:** Liquid scintillation counting; Standardization; TDCR

## 1. Introduction

The triple-to-double coincidence ratio (TDCR) method (Pochwalski et al., 1988; Broda and Pochwalski, 1992) of liquid scintillation (LS) counting continues to be a popular technique used by national metrology laboratories for the standardization of pure  $\beta$ -emitting radionuclides. The National Institute of Standards and Technology (NIST) has recently constructed a TDCR spectrometer to complement measurements currently being carried out using the CIEMAT/NIST  $^3\text{H}$ -efficiency tracing method (Coursey et al., 1986; Zimmerman and Collé, 1997). In order to ensure proper operation and reliability of the newly constructed system, a series of tests was required in which TDCR measurements were carried out in parallel with and compared to assays using other, accepted, techniques. This paper describes

the new NIST TDCR spectrometer and reports the results of those comparisons.

The choice of radionuclides was based primarily on the availability of the radioactive solutions, as well as on the availability of an acceptable technique against which the results could be judged. For the tests described in this paper, solutions of  $^3\text{H}$ ,  $^{54}\text{Mn}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Y}$ ,  $^{103}\text{Pd}$ , and  $^{204}\text{Tl}$  were measured in the TDCR system and compared to a combination of previously calibrated values or results from measurements from LS counting using the CIEMAT/NIST efficiency tracing method, from isothermal calorimetry, from integral  $\beta$ -counting (with extrapolation), or from  $4\pi$   $\beta$ - $\gamma$  anticoincidence counting.

## 2. Experimental

### 2.1. The NIST TDCR system

The design of the NIST TDCR system is based on the systems currently in use in the laboratories at the Laboratoire National Henri Becquerel (LNHB) and

\*Corresponding author.

E-mail address: [b.e.zimmermann@iaea.org](mailto:b.e.zimmermann@iaea.org)  
(B.E. Zimmerman).

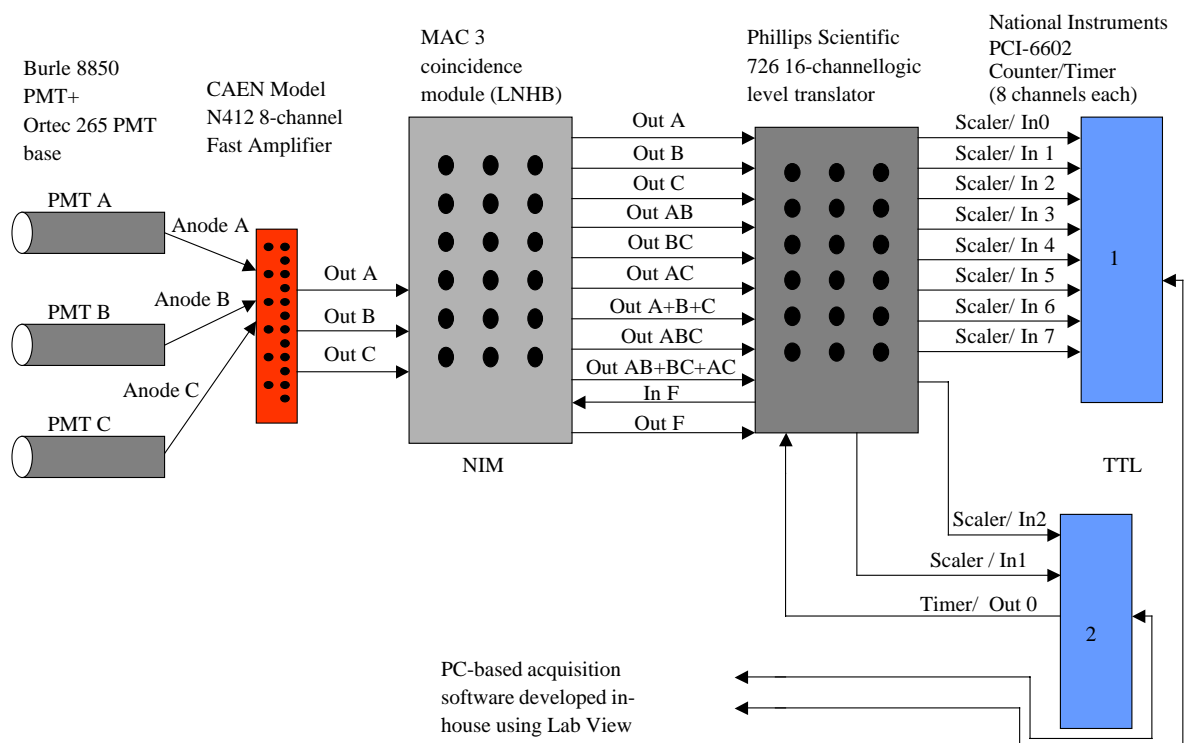


Fig. 1. Schematic of acquisition hardware comprising the TDCR detection system installed at NIST.

POLATOM and is depicted schematically in Fig. 1. The three photomultiplier tubes (PMTs) were modified by reversing the high-voltage polarity and applying it to the anode instead of the photocathode (thereby reducing thermal noise), by adding a high-voltage input to the first dynode (permitting higher gain), and by adding an additional high voltage lead to the focusing electrode (allowing variable focusing voltage, thereby changing detection efficiency). The respective high-voltage inputs for the anode, first dynode, and focusing electrode from each of the three PMTs are connected at a common point, each with its own power supply, so that the voltage on each input for all three tubes can be simultaneously changed.

The three phototubes are arranged in a sample chamber at an angle of  $120^\circ$  apart at a distance, approximately, 1 cm from the LS sample vial. All surfaces of the inside of the sample chamber have been painted with reflective paint to increase the light collection efficiency. The PMTs and sample chamber are enclosed in a light-tight aluminum box that has an upper chamber to allow for dark storage of counting sources and for manual changing of the sources.

The heart of the system is the MAC-3 unit (Bouchard and Cassette, 2000), which was designed and built at LNHB and which contains all of the coincidence logic

and extending deadtime correction circuitry in a single NIM module.

Counting data are acquired using two National Instruments<sup>1</sup> PCI6602 Counter/Timers, each connected to a BNC-2121 BNC connector block for signal input and output. One channel from one of the units is operated in timer mode with a repetition rate of  $10^3 \text{ s}^{-1}$  to provide an oscillator signal to the MAC-3 for deadtime correction. The counter is capable of operating at rates as high as  $10^6 \text{ s}^{-1}$ , but this degree of precision was deemed unnecessary for the present set of measurements. Counter (scaler) inputs are used to record events in each of the three singles channels, each of the three double coincidence channels, as well as the triple coincidence, sum of singles, logical sum of doubles, and livetime channels. Logic level translation between the MAC-3 (NIM) and the PCI6602 cards (TTL) is achieved using a Phillips Scientific 726 16-channel logic level translator.

<sup>1</sup> Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

All of the data acquisition, as well as the high voltage on the focusing electrodes (controlled via an RS-232 serial interface), is controlled through a LabView program that was developed in-house specifically for this application.

All of the counting data for  $\beta$ -emitters were analyzed using the computer codes TDCR-02B or TDCR-02P, which are modified versions of the TDCR-02 code (Broda et al., 2000). The TDCR-02B program contained only minor changes to the original, but the version TDCR-02P added the ability to calculate the detection efficiencies as a function of the TDCR using the Polya distribution in place of the Poisson distribution. This modification was made because of the fact that the Poisson distribution is apparently not valid for extremely small numbers of photoelectrons, such as the case with low-energy  $\beta$ -emitters (Zimmerman et al., 2003). For the set of present measurements, only those dealing with  $^3\text{H}$  required treatment assuming the Polya distribution.

Counting efficiencies for the electron capture (EC) nuclide  $^{103}\text{Pd}$  were calculated as a function of the TDCR (ratio of triple photon coincidences to the logical sum of double photon coincidences) using the program Pd103.2 (Cassette, 2002), which was written specifically for this radionuclide. An important aspect of this program is the assumption that all three PMT efficiencies are equal. For  $^{54}\text{Mn}$ , a second program, TDCREC (Broda, 2002), was developed that has the advantage of being able to take into account differences in PMT efficiencies.

A  $k\text{B}$  value of  $0.012\text{ cm keV}^{-1}$  was used in all of the efficiency calculations, following the results of a study by Broda et al. (2002).

## 2.2. Performance tests

The measurements described in this paper, with the exception of those on previously standardized sources, were conducted in parallel with ongoing calibration projects carried by this laboratory. This provided the opportunity to compare the TDCR results with those obtained using primary methods.

The generic scheme used to prepare solutions for measurement in the TDCR was the same for each experiment. In general, 10 ml of either Beckman Ready-Safe or Wallac OptiPhase HiSafe III cocktail were added to a series of glass LS vials, followed by the addition of between 20 mg and 100 mg of the respective radioactive solution. In some cases, such as for  $^{54}\text{Mn}$  and  $^{103}\text{Pd}$ , the influence of aqueous fraction of cocktail was studied by the addition of nominally 0.5 to 1 ml of distilled water to the cocktail prior to the addition of the radioactivity. Background blanks were prepared so that they had the same chemical composition, in terms of aqueous fraction and ion concentration, as the cocktails containing the radioactive solutions.

The number of efficiency points (voltages applied to the focusing electrode) performed in each measurement series ranged from 4 to 10, with typically 3 repeated counts taken at each voltage. Live counting times at each data point ranged from 30 to 300 s. When possible, the measurement cycles were repeated, usually after all of the cocktails were counted at least once. The number of repeated cycles, particularly for short-lived radionuclides ( $^{90}\text{Y}$ ,  $^{103}\text{Pd}$ ) was limited since samples must be changed manually.

The first tests of the TDCR system were carried out with solutions containing low-energy  $\beta$ -emitters, namely NIST Standard Reference Material 4226C ( $^{63}\text{Ni}$ ) (National Institute of Standards and Technology, 1995; Zimmerman and Collé, 1997) and a gravimetric dilution of NIST Standard Reference Material 4927F (tritiated water) (National Institute of Standards and Technology (NIST), 2000). As a first check of the ability of the system to measure pure EC radionuclides, a solution of  $^{54}\text{Mn}$  was obtained by NIST and calibrated using the NIST calibrated  $4\pi\gamma$  ionization chamber.

Since the primary purpose of these experiments was to be the first investigation into the performance of the TDCR system and not to perform a full calibration of the solutions, there was no intentional experimental design. Instead, the intent was to merely count the LS sources several times under various conditions and check the resulting activities to see if changes to either the apparatus or analysis and acquisition codes were required. Therefore, not all of the counting data collected was appropriate for activity determination. Possible reasons for exclusion from the data set included interruption of the counting cycle, insufficient counting statistics, or insufficient number of efficiency (voltage) points.

The next set of measurements performed using the TDCR system involved the calibration of a solution containing  $^{90}\text{YCl}_3$ . In this study, cocktails were prepared for counting in the TDCR system as described above. The primary measurements in this experiment were performed with the CIEMAT/NIST method. For this, an additional set of LS cocktails were independently prepared from the same master solution (as well as a set of chemically matched  $^3\text{H}$  cocktails for efficiency tracing) and counted in three commercial LS counters. Details of source preparation and counting protocols will be published separately.

Revisiting the assay of EC radionuclides, the next test of the TDCR system involved the measurement of  $^{103}\text{Pd}$  solutions derived from the chemical digestion of two  $^{103}\text{Pd}$  seeds designed for use in the treatment of prostate cancer, one having an activity level consistent with those normally used in cancer therapy, and the other having the same design but having a much higher activity level. The purpose of this second seed was to allow the higher activity seed to be measured singly in the NIST

isothermal calorimeter, which served as the primary measurement against which the TDCR results would be compared. The solutions were also assayed for massic activity using LS counting and the CIEMAT/NIST efficiency tracing method for further comparison. The TDCR cocktails were, in general, prepared as described above. Details of relationship between these sources, as well as a detailed description of the LS source preparation and calorimeter measurements are given in Collé and Zimmerman (2003). Again, the cocktails measured in the TDCR system were generally prepared as described above.

The latest test of the TDCR system included its use in a recent international comparison of  $^{204}\text{Tl}$  Sources were prepared for counting in the TDCR system, the NIST  $4\pi$   $\beta$ - $\gamma$  anticoincidence system (which was also used for integral  $\beta$  counting with extrapolation to provide an additional result), and LS counting using the CIEMAT/NIST method. The cocktails for counting in the TDCR system were prepared as described above. Due to time limitations, each source was counted only three times for 300 s each, with 5 repetitive measurements at each focusing voltage performed during each counting cycle.

### 3. Results and discussion

The results of all of the initial performance tests of the NIST TDCR system are presented in Table 1, along with the results of the corresponding measurements carried out using accepted, primary techniques.

The results of a typical TDCR counting experiment for  $^{63}\text{Ni}$  using the NIST TDCR spectrometer can be seen in Fig. 2. The first striking characteristic of this instrument is the relatively large inequality of the counting efficiencies between the double coincidence channels despite modest attempts to match the PMTs. Despite this, the programs TDCR-02B and TDCR-02P are able to adequately compensate for this effect.

For  $^3\text{H}$ , using the Polya distribution for the efficiency calculation, the average massic activity,  $C_A$ , was  $28.10 \pm 0.29 \text{ kBq g}^{-1}$ . The uncertainty is an expanded ( $k=2$ ) uncertainty calculated from the quadratic addition of the standard deviation on the activity determinations for typically 8 points in a counting series having different efficiency values (average of 0.09%) and the standard deviation on 5 independent determinations of  $C_A$  with three sources (0.5%). This result is in excellent agreement with the previously certified value of  $28.11 \pm 0.20 \text{ kBq g}^{-1}$ , where the uncertainty is also an expanded ( $k=2$ ) uncertainty.

The average value of  $C_A$  for the  $^{63}\text{Ni}$  solution was measured to be  $48.18 \pm 0.32 \text{ kBq g}^{-1}$ . The uncertainty is an expanded ( $k=2$ ) uncertainty calculated from the quadratic addition of the standard deviation on the

Table 1

Results of initial TDCR measurements and comparisons with previously calibrated values ( $C_{A, \text{cert}}$ ) or with results of measurements using isothermal calorimetry ( $C_{A, \text{calor}}$ ), LS counting with the CIEMAT/NIST efficiency tracing method ( $C_{A, \text{C/N}}$ ), integral  $\beta$ -counting ( $C_{A, \beta, \text{int}}$ ), or  $4\pi$   $\beta$ - $\gamma$  anticoincidence counting ( $C_{A, \text{Anticoin}}$ ). All uncertainties are expanded ( $k=2$ ) uncertainties

Radionuclide	$C_{A, \text{TDCR}}$	$C_{A, \text{Cert}}$	$C_{A, \text{Calor}}$	$C_{A, \text{C/N}}$	$C_{A, \beta, \text{int}}$	$C_{A, \text{Anticoin}}$
$^3\text{H}$	$28.10 \pm 0.29 \text{ kBq g}^{-1}$	$28.11 \pm 0.20 \text{ kBq g}^{-1}$	—	—	—	—
$^{54}\text{Mn}$	$31.56 \pm 2.28 \text{ kBq g}^{-1}$	$33.24 \pm 23 \text{ kBq g}^{-1}$	—	—	—	—
$^{63}\text{Ni}^a$	$48.18 \pm 0.32 \text{ kBq g}^{-1}$	$48.26 \pm 0.44 \text{ kBq g}^{-1}$	—	—	—	—
$^{90}\text{Y}$	$11.17 \pm 0.17 \text{ MBq g}^{-1}$	—	—	$11.09 \pm 0.10 \text{ MBq g}^{-1}$	—	—
$^{103}\text{Pd}^b$	$9.54 \pm 1.43 \text{ MBq g}^{-1}$	—	$91.46 \pm 1.46 \text{ MBq g}^{-1}$	$93.68 \pm 1.59 \text{ MBq g}^{-1}$	—	—
	$1.672 \pm 0.025 \text{ MBq g}^{-1}$	—	$1.612 \pm 0.029 \text{ MBq g}^{-1}$	$1.638 \pm 0.038 \text{ MBq g}^{-1}$	—	—
$^{204}\text{Tl}$	$101.83 \pm 0.92 \text{ kBq g}^{-1}$	—	—	$102.22 \pm 0.94 \text{ kBq g}^{-1}$	$102.07 \pm 1.26 \text{ kBq g}^{-1}$	$101.82 \pm 0.92 \text{ kBq g}^{-1}$

<sup>a</sup> The Standard Reference Material, SRM 4226C, used in the preparation of the TDCR LS cocktails for these measurements was calibrated in 1995 by LS counting with the CIEMAT/NIST efficiency tracing method (Zimmerman and Collé, 1997).

<sup>b</sup> Two different solutions of  $^{103}\text{Pd}$  were used in the measurements, each prepared from a chemical digestion of two different activity-level  $^{103}\text{Pd}$  seeds. The seeds from one of these levels were separately assayed in the isothermal calorimeter prior to digestion and were gravimetrically linked to its subsequently prepared master solution. The two solutions in turn were linked through relative measurements by LS counting, ionization currents, and photon spectrometry.

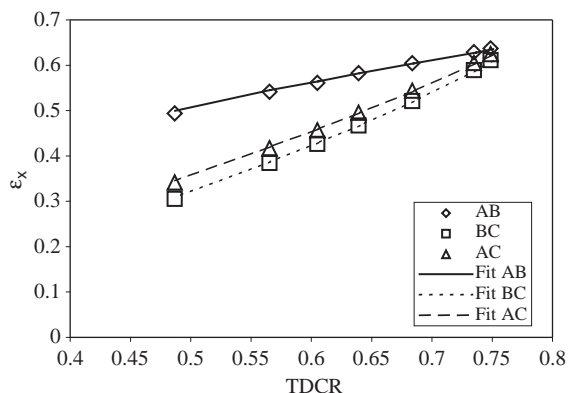


Fig. 2. Experimental and theoretical counting efficiencies,  $\varepsilon_x$ , for double coincidence channels AB, BC, and AC as a function of the ratio of triple-photon to double-photon coincidences (TDCR) for a single  $^{63}\text{Ni}$  LS cocktail. Each point represents two repeated counts at each focusing voltage (efficiency value), with the standard deviation of each measurement lying within the respective symbol for each point. The theoretical efficiencies (lines) were calculated with the program TDCR-02B using a Poisson distribution and a  $k_B$  value of  $0.012 \text{ cm keV}^{-1}$ .

activity determinations for typically 8 points in a counting series having different efficiency values (average of 0.11%) and the standard deviation on 4 independent determinations of  $C_A$  with two sources (0.33%). Again, this result is in excellent agreement with the previously certified massic activity value of  $48.26 \pm 0.44 \text{ kBq g}^{-1}$ , where the uncertainty is an expanded ( $k=2$ ) uncertainty.

For the  $^{54}\text{Mn}$  solution, data from counts on 7 different sources (two having repeated cycles) were available for analysis. From these data, an average massic activity of  $31.56 \pm 2.28 \text{ kBq g}^{-1}$  was obtained. The uncertainty is an expanded ( $k=2$ ) uncertainty. In these measurements, the largest uncertainty component was that due to the variability in activity values calculated at each efficiency point by the TDCREC program (3.6%). This was combined with the variability between activity determinations for each source (0.57%) to give a combined relative uncertainty of 3.64%.

Although there is agreement with the calibrated value of  $33.24 \pm 0.23 \text{ kBq g}^{-1}$  due to the large uncertainty on the  $^{54}\text{Mn}$  TDCR measurement, the relative difference between the two values is 5.3%. This highlights the difficulty in measuring this nuclide. The PMT asymmetry is further exacerbated by the relatively low detection efficiency of the NIST system compared to those at POLATOM and LNHb. In the case of the present experiments, a maximum doubles counting efficiency of about 24% for  $^{54}\text{Mn}$  was achieved.

A total of 11 cocktails were counted in the TDCR system, each at least twice, for the experiments involving

$^{90}\text{Y}$ . From these data, an average massic activity of  $11.17 \pm 0.17 \text{ MBq g}^{-1}$  was determined for the master solution, compared to  $11.09 \pm 0.10 \text{ MBq g}^{-1}$  from the CIEMAT/NIST efficiency tracing data. The uncertainties on both results are expanded ( $k=2$ ) uncertainties. As was the case for  $^{54}\text{Mn}$ , the variability in the activity calculated at each activity point within a measurement cycle was the largest component of uncertainty, with a magnitude of 0.4%. The other large components included variability due to reproducibility between independently prepared cocktails (0.26%), measurement repeatability on the same cocktail (0.15%), the uncertainty in the dilution factor from the master solution (0.23%), and the uncertainty in the  $^{90}\text{Y}$  half-life (0.14%). On a relative basis, the TDCR and CIEMAT/NIST results differ by 0.7%, but agree to be within the experimental uncertainties.

The measurements of  $^{103}\text{Pd}$  represented the first-ever standardization of this radionuclide by NIST. For this reason, measurements were carried out using as many different techniques as possible. While the details of the study are beyond the scope of this paper (and will be published separately), it should be noted that an extraordinarily large set of measurements went into the results presented in Table 1. Because there were two seeds that were independently digested and measured, there were two independently prepared solutions (with differing activity levels) available to assay. In all, data were acquired in the TDCR system using seven LS cocktails with each of the  $^{103}\text{Pd}$  solutions (for a total of 14), four of them having been counted at least twice.

An important aspect of the TDCR data analysis was the fact that the program that was used to calculate the doubles counting efficiencies as a function of TDCR assumes that the efficiencies of the PMTs are equal and that the efficiencies of all three tubes change equally as a function of the applied focusing voltage. This was found to be the case only when the focusing voltage,  $V_f$ , was greater than or equal to 760 V. Physical characteristics of the phototubes limited the maximum allowable  $V_f$  to 795 V, which means that there is only a possible 35 V range over which the focusing can be changed. Because of the high detection efficiency for  $^{103}\text{Pd}$ , it is rather insensitive to such small changes in the focusing voltage. In the present study, a typical maximum change in detection efficiency over that range was less than 1.5%. Because of the requirement imposed by the model, only data acquired in the voltage range 760–795 V were used in computing the mean massic activity.

From the data in Table 1, it is evident that the results from both of the LS techniques are higher than those from calorimetry, suggesting that the LS methods underestimate the efficiency. Recent evaluations of the  $^{103}\text{Pd}$  level scheme also suggest that the branching ratio



for the isomeric state may be in error by as much as 2% (Collé and Zimmerman, 2003). Because the calorimeter measures total absorbed power, it is likely to be much less sensitive to errors in the branching ratio than would LS counting, in which the relative abundances of electrons (with relatively high counting efficiencies) and photons (with somewhat lower efficiencies) can greatly affect the calculated efficiency. The two LS techniques are in agreement to within their respective measurement uncertainties, as are the CIE-MAT/NIST and calorimetry results. However, there is no overlap in the TDCR and calorimetry results. In light of these data and the suspect nature of the level scheme, it is clear that more research is necessary to resolve these issues.

Perhaps the most gratifying results to date are those from the recent international comparison of  $^{204}\text{Tl}$ . As seen in Table 1, the activity values for the  $^{204}\text{Tl}$  solution obtained for all four measurement methods agreed to within the respective experimental uncertainties. In addition, no two results differed by more than 0.39%, and the mean combined standard uncertainty for the 4 techniques was 0.5%. For the TDCR measurements, the primary uncertainty component was the measurement reproducibility (0.4%). Because of time constraints, insufficient data were acquired in the TDCR system to fully analyze for other components usually identified as being significant, such as measurement repeatability and cocktail composition.

One possible reason for the success achieved for this radionuclide with the TDCR system was the high detection efficiency for  $^{204}\text{Tl}$  and the ability to use an analysis code that takes into account the PMT asymmetry. Typical counting efficiencies during these measurements were about 95% and, as in the case of  $^{103}\text{Pd}$ , the efficiency did not change significantly over the range of focusing voltages used.

#### 4. Conclusion

The results of these tests of the NIST TDCR system indicate that despite the significant asymmetry in the efficiencies of the three PMTs, it is capable of providing accurate activity measurements for beta emitters as long as the data are analyzed with a model that accounts for such differences in PMT response. Several areas for improvement still exist, however, for the case of electron capture nuclides and their measurement with the system, primarily due to the low intrinsic detection efficiency and the lack of a generally applicable analysis program.

#### Acknowledgements

The authors gratefully acknowledge the advice and assistance in constructing the NIST TDCR system given by Drs. Ryszard Broda (POLATOM) and Philippe Cassette (LNHB), without whose help construction of the NIST TDCR system would not have been possible.

#### References

- Bouchard, J., Cassette, P., 2000. MAC3: an electronic module for the processing of pulses delivered by a three photo-multiplier liquid scintillation counting system. *Appl. Radiat. Isot.* 52, 669–672.
- Broda, R., 2002. Private communication.
- Broda, R., Pochwalski, K., 1992. The enhanced triple to double coincidence ratio (EDTCR) method for standardization of radionuclides by liquid scintillation counting. *Nucl. Instrum. Methods A* 312, 85–89.
- Broda, R., Cassette, P., Maletka, K., Pochwalski, K., 2000. A simple computing program for application of the TDCR method to standardization of pure beta emitters. *Appl. Radiat. Isot.* 52, 673–678.
- Broda, R., Maletka, K., Terlikowska, T., Cassette, P., 2002. Study of the influence of the LS-cocktail composition for the standardization of radionuclides using the TDCR method. *Appl. Radiat. Isot.* 56, 285–289.
- Cassette, P., 2002. Private communication.
- Collé, R., Zimmerman, B.E., 2003. Primary microcalorimetric standardization of  $^{103}\text{Pd}$ , its comparison to LS spectrometry, and its application to the calibration of seeds used for prostate-cancer therapy. In preparation.
- Coursey, B.M., Mann, W.B., Grau Malonda, A., Garcia-Toraño, E., Los Arcos, J.M., Gibson, J.A.B., Reher, D., 1986. Standardization of Carbon-14 by  $4\pi\beta$  liquid scintillation efficiency tracing with hydrogen-3. *Appl. Radiat. Isot.* 5, 403–408.
- National Institute of Standards and Technology (NIST), 1995. Certificate, standard reference material 4226C, radioactivity standard, Nickel-63. Gaithersburg, MD.
- National Institute of Standards and Technology (NIST), 2000. Certificate, standard reference material 4927F, radioactivity standard, Hydrogen-3. Gaithersburg, MD.
- Pochwalski, K., Broda, R., Radoszewski, T., 1988. Standardization of pure beta emitters by liquid scintillation counting. *Appl. Radiat. Isot.* 39, 165–172.
- Zimmerman, B.E., Collé, R., 1997. Standardization of  $^{63}\text{Ni}$  by  $4\pi\beta$  liquid scintillation spectrometry with  $^3\text{H}$ -standard efficiency tracing. *J. Res. Natl. Inst. Stand. Technol.* 102, 455–477.
- Zimmerman, B.E., Collé, R., Cessna, J.T., Broda, R., Cassette, P., 2003. Application of the triple-to-double coincidence ratio method at NIST for absolute standardization of radionuclides by liquid scintillation counting. *ACS Symp. Ser.* 868 (Radioanalytical Methods in Interdisciplinary Research), Love, C.A., Nash, K.L., eds., pp. 76–87.