Report of Test

for

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Palladium-103 Activity Calibrations of "TheraSeed" Brachytherapy Sources

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I. Submitted sources

At the request of Theragenics Corporation (Buford, GA), two batches of sealed sources of ^{103}Pd were submitted for activity calibrations and were received on 6 May 2002. The first batch of three sources, were transmitted through the NIST Dosimetry Group and were identified as being "TheraSeed Model 200 Palladium-103" sources, each having a reported "apparent activity" (sic) of 2.63 mCi at a reference time of 1200 EST 29 April 2002. They were designated in each case as being "Hearn Test Seeds (Order O105154), Lot ID 0215H, Product TG202" and further identified with labels of "S60", "S61" and "S62." The second batch of seeds, received directly from Theragenics, consisted of eight sources that were said to have been manufactured such that they were essentially identical to the first batch of sources, excepting that they were specially-fabricated to contain elevated activity levels (as well as having the possibility of containing a different relative quantity of palladium carrier). They had identification labels of "HC-1" through "HC-8" and each was reported to have an "activity" of approximately 220 mCi at a reference time of 2 May 2002. Table 1 summarizes this information on the submitted sources, along with corresponding NIST-assigned identity labels.

Table 1. Submitted sources.

Batch identification by Theragenics	Number of sources in batch	Reported activity in each source	Reported reference time	NIST assigned identity labels
S60 through S61	3	2.63 mCi ("apparent")	1200 EST 29 April 2002	L1 through L3, respectively
HC-1 through HC-8	8	approximately 220 mCi	2 May 2002	H1 through H8, respectively

The NIST calibration measurements (excluding data analyses and reconciliation) were initiated on 6 May 2002 and were completed on 12 August 2002.

A summary of the certified values, as taken from subsequent Tables 8 and 11, for the ^{103}Pd activity in each of the eleven submitted sources, at a reference time of 1200 EST 6 May 2002, follows:

source identity	^{103}Pd activity (GBq)	relative expanded ($k = 2$) uncertainty (in %)
H1	5.862	
H2	5.869	
H3	5.941	
H4	5.927	
H5	5.847	1.7
H6	5.890	
H7	5.834	
H8	5.882	
L1	0.1347	
L2	0.1347	1.9
L3	0.1334	

II. Overview of calibration measurements

Calorimetric measurements as a function of decay time were performed to obtain the total power from six of the high-level sources, namely H2 through H7. Five independent trials were conducted, under varying source treatments and containment conditions. Each trial consisted of continuous power measurements (with independent power calibrations) over decay time intervals ranging from 7.4 to 24.3 days.

These power measurements were converted to total ^{103}Pd activities through the use of a calculated average energy per decay whose value was based on nuclear-decay data.

Relative ionization current responses on all 11 seeds, as obtained from two different instruments in standard source geometries, were then used to assign an individual activity to each source from the relative proportion of the total calorimetrically-determined activity.

Direct comparisons between the low- and high-level sealed sources were precluded because of possible differences in their external spectra that might result from differences in the ratio of palladium carrier to active ^{103}Pd in the two source batches. Therefore, to link the low-level sources (L1 through L3) to the high-level sources (H1 through H8), sources L2 and H8 were radiochemically digested. The resulting solutions ML and MH were assayed, for confirmatory purposes, by liquid scintillation (LS) spectrometry using both a CIEMAT/NIST efficiency-tracing (LSET) approach (using a standardized ^{54}Mn solution as the tracer) and by a triple-to-double-coincidence ratio (TDCR) method. The two solutions were also intercompared, to link the sources, by relative ionization current measurements, by relative high-resolution photonic-emission spectrometry, and by relative LS counting.

III. Assumed nuclear decay data

Relevant nuclear data, as obtained from a recent BNM-CEA/LNHB re-evaluation [1], which was used to convert calorimetric power into activity, is provided in Table 2.

Table 2. Nuclear decay and atomic data for the average energy per decay and the power (in units of μW) per unit activity (in units of GBq).

nuclide	average energy per decay (keV)	power per unit activity (μW$\cdot$GBq^{-1})	half-life
^{103}Pd	60.82 $\pm$ 0.08	9.744 $\pm$ 0.013	16.991 $\pm$ 0.019 d

The above cited uncertainty in the average energy per decay (0.13 % on a relative basis) is based exclusively on the tabulated data for the extant decay scheme. Inconsistencies in this decay scheme, however, suggest that the value for the average energy per decay could be in error by as much as 1 % to 2 % with renormalizations.

This BNM-CEA/LNHB data re-evaluation for ^{103}Pd was also used for the LS-based TDCR and efficiency tracing LSET calculations, while the data for ^{54}Mn were taken from ENSDF [2].

IV. Microcalorimetry on the H-level sources

Calorimetric power measurements were made with a commercial dual-cell, near-isothermal (heat flow) calorimeter that was obtained from Calorimetry Sciences Corporation (CSC) of Spanish Fork, Utah and which was denoted as "CSC 4400 Isothermal Microcalorimeter (IMC)." The calorimeter was suitably adapted to utilize specially-designed and -fabricated source-holder cells that are used to maximize the energy absorption of the ionizing radiation and incorporates resistance heaters within these measurement cells to obtain very-accurately-determined independent power calibrations. Refer to Collé and Zimmerman [3] for details on the calorimeter, its operation, and performance evaluations.

Measurements were performed with both thick-walled brass and aluminum source-holder energy absorbers within Hastelloy-C cells. Monte Carlo calculations for the energy deposition in the measurement cells were used to assess and correct for the power loss due to escaping ionizing radiation. The calculated fractions, as lower bounds, of the total energy absorbed within the sources and cells were 0.9995 for the brass absorber and 0.9982 for the aluminum absorber.

Each calorimetric determination, for each of five independent insertion trials, was based on temporal measurements of the power obtained with the batch of six sources in a sample cell S (as measured against comparable blank materials in a matched reference cell R). The power P_0 from the sources at a reference time $t = 0$ was obtained by fitting the power decay data (P as a function of time t) to the functional form $P(t) = P_0\exp(-\lambda t) + P_B$ where λ is the well-known decay constant for ^{103}Pd [$\lambda = \ln(2)/16.991$ d^{-1}] and where P_B is an average baseline power that corresponds to the null difference between the S and R cells in the absence of a source. All power $P(t)$ measurements were determined at near-isothermal conditions at steady-state heat flow.

The power measurements were obtained using an average "canonical" power calibration factor that was determined throughout the course of this work. Thirteen independent heat-flow power calibrations, at applied powers ranging from 30 μW to 250 μW, were performed by applying known power levels to both the S and R cells under both baseline and inserted source conditions. At least two calibrations were performed for each insertion trial.

Following the first three trials, two insertions in brass and one in aluminum, two further trials were conducted to evaluate potential source-composition "heat defect or excess" thermochemical effects. Firstly, the six sources were heated to 565 °C for 2.5 hours to alter the internal source structure prior to re-insertion in aluminum for a fourth trial. Secondly, for the fifth insertion trial, the sources were enclosed within a graphite sleeve inside the brass absorber to determine if there was a radiation-on-graphite thermal effect. The calorimeter was restored to baseline conditions between each insertion trial.

The results from the five independent insertion trials used to obtain the calorimetric power and the equivalent ^{103}Pd activity for the six H-level sources are given in Table 3. Uncertainty assessments for both the calorimetric power and equivalent ^{103}Pd activity are given in Table 4. **Certified values are in bold font.**

Unless specifically noted otherwise, all uncertainties and uncertainty intervals cited within this Report correspond to derived "standard uncertainties" or propagated "combined standard uncertainties." Certified values are reported in terms of "expanded uncertainties" with "coverage factors" of $k = 2$ (see references [4] and [5]).

Table 3. Total calorimetric power and equivalent ^{103}Pd activity for the six H-level sources at a Reference Time of 1200 EST 6 May 2002. **Certified values are in bold font.**

Insertion trial	Insertion conditions	Measurement interval (days)	Power (μW) †	Activity (GBq) †
1st	In brass absorber	7.4	343.05 ± 2.11	35.205 ± 0.217
2nd	Re-inserted into brass	9.2	345.26 ± 1.71	35.432 ± 0.175
3rd	In aluminum absorber	12.7	343.70 ± 0.99	35.272 ± 0.102
4th	After firing, re-inserted into aluminum	24.3	343.72 ± 0.30	35.274 ± 0.031
5th	With excess graphite, re-inserted into brass	18.4	344.60 ± 0.47	35.295 ± 0.048
MEAN			**344.07**	**35.309**
Relative standard deviation of the mean			**0.87**	**0.089**

† The cited uncertainties correspond to standard deviations of the mean and were derived exclusively from the propagated statistical uncertainty on the fitted P_0 .

Table 4. Assessment of the uncertainty in the total calorimetric power and equivalent ^{103}Pd activity for the six H-level sources as given in Table 3. **Certified values are in bold font.**

Uncertainty component	relative standard uncertainty (in %)	
	Power	Activity
Standard deviation of fitted calorimetric power for single insertion; median value (Table 3)	0.29	
Measurement precision for five replicate insertions; propagated standard deviation of the mean (Table 3)	0.12	
Calorimetric baseline stability, radionuclidic impurity contributions, and fitted model assumptions	0.3	
Radiative energy loss from Monte Carlo calculations	< 0.1	
"heat defect or excess" thermochemical effects	0.5	
Power calibration accuracy (single determination)	< 0.05	
Calorimeter calibration factor; standard deviation of the mean for n = 13 determinations	0.06	
^{103}Pd decay corrections over measurement intervals	0.21	
Calorimetric power combined standard uncertainty	0.71	--
Expanded uncertainty (k = 2) in total calorimetric power	**1.4**	--
Average energy per decay	--	0.13
^{103}Pd activity combined standard uncertainty	--	0.72
Expanded uncertainty (k = 2) in total ^{103}Pd activity	--	**1.4**

V. Ionization current measurements on the individual sources

Ionization current measurements on all 11 submitted sources were performed with two different instruments: (1) NIST chamber "A", a unique, argon-filled, pressurized (2 MPa) "$4\pi\gamma$" re-entrant ionization chamber (hereafter designated as PIC "A"); and (2) a Vinten Instruments Ltd. (Surrey, UK) model 671, nitrogen-filled, pressurized (1 MPa) re-entrant ionization chamber (designated as PIC "V").

Current measurements with PIC "A" were obtained with respect to NIST, internal-standard, sealed ^{226}Ra reference sources, RRS3, RRS10, and RRS20 and are reported in terms of background- and decay-corrected current ratios $\Gamma = I_i / I_R$ at a common reference time of 1200 EST 6 May 2002 (where I_i is the corrected current for source i and where I_R is that for RRS reference source.

Current measurements with PIC "V" are reported directly in terms of negative-potential currents (in pA) that were corrected for background and radioactive decay to the 1200 EST 15 December 2001 reference time. A check source of ^{226}Ra (nominal 9.3 pA) was used to assure instrument consistency between measurement occasions.

For the ionization current measurements with both instruments, the individual sources were contained within a specially-fabricated, source-holder insert that just fitted within both instruments' standard-ampoule source holder. The insert consisted of a "Lucite" (methyl methacrylate) right-circular cylinder rod with a 1.5- mm inner diameter and a 2.0-mm wall and whose external dimensions (76-mm length, 16.5-mm outer diameter, and 65-mm well depth) -- and hence source positioning -- matched that for the standard-ampoule source holders.

Average ionization current ratios (for PIC "A") and ionization currents (for PIC"V") were derived from numerous multiple measurements of ionization currents, including evaluations of source positioning variability, on either two or three separate measurement occasions over the course of 6 days. The averaged results for both instruments are given in Table 5, along with estimates of the replicate measurement precision on each.

Table 5. Results of the ionization current measurements on the individual sources at the reference time of 1200 EST 6 May 2002. The results are given in terms of the mean ionization current ratios Γ for PIC "A" and the mean ionization current I (in pA) for PIC "V". The measurement precision on each mean is given in terms of the propagated relative standard deviation of the mean s_m.

	PIC "A"				PIC "V"	
source identity	mean Γ (RRS3)	mean Γ (RRS10)	mean Γ (RRS20)	s_m / Γ (%)	mean I (pA)	s_m / I (%)
L1	0.17990	0.053700.	--	0.20	8.463	
L2	0.17994	0.053702	--	0.088	8.454	1.2
L3	0.17819	0.053179	--	0.082	8.469	
H1	--	2.2430	0.90013	0.043	365.9	
H2	--	2.2458	0.90126	0.12	371.8	
H3	--	2.2746	0.91228	0.060	371.6	
H4	--	2.2676	0.91003	0.059	372.2	0.23
H5	--	2.2373	0.89786	0.10	362.2	
H6	--	2.2536	0.90441	0.095	368.9	
H7	--	2.2323	0.89585	0.081	359.5	
H8	--	2.2507	0.90321	0.072	368.8	

Correlation analyses demonstrate that the two sets of ionization current measurements within either the L-level or H-level batch exhibit excellent consistency.

VI. Derived ionization-chamber calibration factors for the H-level sources

Calibration factors for the H-level sources for both ionization chambers were derived by summing the ionization currents for all six sources (H2 through H8) used for the calorimetric measurement and combining this result with the calorimetrically-determined equivalent ^{103}Pd activity (Table 3). The results, given as $C_V = \Sigma_i I_i / A$ for PIC "V" and $C_A = \Sigma_i \Gamma_i / A$ for PIC "A", are given in Table 6. Table 7 contains the uncertainty assessment for the derived calibration factors.

Table 6. Derived calibration factors for the PIC "V" and PIC "A" ionization chambers at a Reference Time of 1200 EST 6 May 2002.

sources	$\Sigma_i I_i$ (pA)	$\Sigma_i \Gamma_i$ (RRS20)	^{103}Pd activity A (GBq)	$C_V = \Sigma_i I_i / A$ (pA $\cdot$GBq^{-1})	$C_A = \Sigma_i \Gamma_i / A$ (GBq^{-1})
H2-H7	2206.1	5.4217	35.309	62.48	0.1536

Table 7. Uncertainty assessment for the derived calibration factors given in Table 6.

sources	relative standard uncertainties (in %)				
	$\Sigma_i I_i$	$\Sigma_i \Gamma_i$	^{103}Pd activity A	$C_V = \Sigma_i I_i / A$	$C_A = \Sigma_i \Gamma_i / A$
H2-H7	0.56	0.22	0.72	0.91	0.75

VII. Assigned activity values on the individual H-level sources

Table 8 contains the assigned activities on the individual H-level sources as determined from the individual mean ionization current ratios Γ for RRS20 (Table 5) from the PIC "A" chamber and its calorimetrically-determined C_A calibration factors (Table 6).

Table 8. Assigned values of the ^{103}Pd activity for the individual sources at a Reference Time of 1200 EST 6 May 2002. **Certified values are in bold font.**

source identity	^{103}Pd activity (GBq)	relative expanded ($k = 2$) uncertainty (in %)
H1	**5.862**	
H2	**5.869**	
H3	**5.941**	
H4	**5.927**	
H5	**5.847**	**1.7**
H6	**5.890**	
H7	**5.834**	
H8	**5.882**	

The certified activity values in Table 8 assume that there are no radionuclidic impurities in the sources that could have significantly contributed to either the calorimetric power measurements or to the ionization currents.

The relative expanded uncertainties for a coverage factor of $k = 2$ on the individual activity values, as given in Table 8, were propagated from the standard uncertainties on the calorimetrically-determined PIC "A" calibration factors (Table 7) and on the decay- and background-corrected PIC "A" relative ionization current measurements. The uncertainty includes contributions due to within- and between-measurement-occasion precision, source-positioning variability, long-term temporal variability in the RRS20 relative ionization current measurements, charge collection effects, livetime determinations, background subtractions, and decay corrections.

There are no apparent statistically-significant differences amongst the activity in the eight H-level sources. Their distribution was tested for normality and they can be considered to have been sampled from a distribution having a mean of about 5.88 GBq and with a standard deviation of about 0.04 GBq, which corresponds to a relative standard deviation of the mean of about 0.2 %.

VIII. Radioanaytical digestion of sources L2 and H8

Source L2 was initially digested in a small volume of a strong acid solution (nominally 80 % concentrated HF and 20 % concentrated HNO_3). After the stainless steel encapsulation was dissolved, the remaining source debris (i.e., mainly the two charcoal pellets) was sequentially leached with an acidic palladium carrier solution. All digestion and leaching solutions were quantitatively transferred and subsequently diluted (serially) to form a master solution ML having a massic activity of about 2 MBq·g^{-1} in 83.576 g of approximately 1 mol·L^{-1} HCl with carrier ion concentrations of about 50µg Pd^{+2} per gram of solution. The total residual activity in the source-dissolution vessel and on all solution-transfer tools, as determined by direct LS measurements, was found to be (0.004 ± 0.002) % of the total activity contained in source L2. This residual activity determination was also confirmed by relative ionization chamber measurements. The kind of quantitative procedures and techniques that were employed for this destructive assay have been described in a generalized way previously (see Collé [6] and references therein).

Subsequent serial dilutions of master solution ML exhibited inhomogeneities that have been attributed to the presence of microscopically-sized pieces of charcoal-based source debris in these solutions. It is assumed that this resulted from partial destruction of the charcoal pellets because of excessive leaching during the digestion procedure. The existence of these solution inhomogeneities resulted in the need to perform relative assays on over thirty-two different solution aliquants and dilutions. It was found that the solution inhomogeneities were mainly present within the dilutions of ML and not in aliquants of ML itself.

Source H8 was digested in a manner similar to that employed for source L2, excepting that the leaching was deliberately terminated at an earlier stage to avoid destruction of the charcoal-pellet debris. In this case, the residual activity was found to be (0.12 ± 0.04) % of the total activity contained in source H8, which corresponded to an activity that could not be directly measured by LS counting until after nearly three months of decay. This residual activity determination was confirmed by relative ionization current measurements in three different commercial re-entrant ionization chambers and by gross photon counting with a NaI(Tl) well counter. The resulting master solution MH had a massic activity of about 100 MBq·g^{-1} in 69.238 g of solution (approximately 1 mol·L^{-1} HCl with carrier ion concentrations of about 50µg Pd^{+2} per gram of solution). Aliquants of solution MH and subsequent serial dilutions did not exhibit any inhomogeneity.

IX. Intercomparisons of master solutions ML and MH to link the activity in source L2 to H8

The relative massic activity for the two master solutions ML and MH, in terms of the ratio MH/ML, was determined by relative LS counting and found to be **MH/ML = 56.737 ± 0.027**. The cited uncertainty interval is a standard deviation of the mean for a grand average having a minimum of $(10x2x3 -1) = 59$ degrees of freedom, which corresponds to 0.05 % on a relative basis. The relative comparisons were performed using ten different combinations of ML and MH aliquants in LS cocktails of two different compositions (four vials for each aliquant and composition). Each LS vial was measured in all three of the Group's LS spectrometers on either one or two separate measurement occasions.

Confirmatory measurements for the MH/ML ratio were also obtained from: (i) relative LS counting by LNHB [7], which was performed as part of an international intercomparison, (ii) by x-ray spectrometry of point sources prepared from master solutions ML and MH; and (iii) by relative ionization current measurements with three commercial re-entrant chambers of 5 g aliquants of solutions ML and MH that were put up in standard NIST 5-mL flame-sealed ampoules.

These confirmatory measurements had relative massic activity values of:
- (i)   LNHB LS counting   MH/ML = 56.85 ± 0.05   (or +0.20 % different);
- (ii) x-ray spectrometry   MH/ML = 56.67 ± 0.12   (or -0.12 % different); and
- (iii) ionization currents   MH/ML = 57.1 ± 0.2   (or +0.6 % different)

where the cited uncertainty intervals represent expanded ($k = 2$) standard uncertainties.

From the calorimetrically-determined activity for source H8 (Table 8) and the massic activity ratio MH/ML for master solutions MH and ML, with normalizations by the total master solution masses and corrections for the residual activities, the activity of source L2 was found to be 134.7 MBq at the 1200 EST 6 May 2002 reference time.

The ratio of the calorimetrically derived activities for sources H8 and L2 was H8/L2 = 43.67; whereas the ratio derived from direct PIC "A" ionization current measurements for the two sealed sources was H8/L2 = 41.91. The 4.1 % difference in the two ratios is most likely due to differences in the external spectra for the H-level and L-level sources.

X. Derived ionization-chamber calibration factors for the L-level sources

Calibration factors for the L-level sources for both ionization chambers were derived from the measured ionization currents (Table 5) and the linked calorimetrically-based activity of source L2. The results, given as $C_V = \sum_i I_1 / A$ for PIC "V" and $C_A = \sum_i \Gamma_1 / A$ for PIC "A", are given in Table 9. Table 10 contains the uncertainty assessment for the derived calibration factors.

Table 9. Derived calibration factors for the PIC "V" and PIC "A" ionization chambers at a Reference Time of 1200 EST 6 May 2002.

source	I_1 (pA)	Γ_1 (RRS3)	^{103}Pd activity A (MBq)	$C_V = \sum_i I_1 / A$ (pA $\cdot$ kBq^{-1})	$C_A = \sum_i \Gamma_1 / A$ (kBq^{-1})
L2	8.463	0.17994	134.7	62.83	1.336

Table 10. Uncertainty assessment for the derived calibration factors given in Table 9.

source	relative standard uncertainties (in %)				
	I_1	Γ_1	^{103}Pd activity A	$C_V = \sum_i I_1 / A$	$C_A = \sum_i \Gamma_1 / A$
L2	1.2	0.2	0.8	1.4	0.8

XI. Assigned activity values on the individual L-level sources

Table 11 contains the assigned activities on the individual L-level sources as determined from the individual mean ionization current ratios Γ for RRS3 (Table 5) from the PIC "A" chamber and its calorimetrically-determined C_A calibration factor (Table 10).

Table 11. Assigned values of the ^{103}Pd activity for the individual sources at a Reference Time of 1200 EST 6 May 2002. **Certified values are in bold font.**

source identity	^{103}Pd activity (MBq)	relative expanded ($k = 2$) uncertainty (in %)
L1	**134.7**	
L2	**134.7**	1.9
L3	**133.4**	

The certified activity values in Table 11 assume that there are no radionuclidic impurities

in the sources that could have significantly contributed to either the calorimetric power measurements or to the ionization currents.

XII. Confirmatory results from LS spectrometry

Primary standardizations of [103]Pd by two LS methods were attempted for comparisons with the calorimetrically-based results. For the first, the master solutions ML and MH were assayed for [103]Pd activity by an efficiency tracing LS spectrometric method (denoted LSET) that has become routine in this laboratory. It is based on well-established procedures for the assay of gravimetrically-determined aliquants of solutions containing nuclides that decay by electron capture (EC) or β-emission (see Collé and Zimmerman [8] and references therein). The assays were performed with 16 series of LS cocktail samples, each with between 3 and 5 independently-prepared sample vials, using three different commercially-available scintillation fluids. Each series of the prepared cocktails was measured in all three of the Group's LS spectrometers. The [103]Pd detection efficiencies were calculated using a modified version of the computer program *EMI* [9] with [54]Mn as the efficiency monitor. The [54]Mn -standard cocktails used for the efficiency tracing were prepared with aliquants of a gravimetric dilution of a [54]Mn standard solution, which was specifically calibrated by NIST for this work. Efficiency variations for the tracing were achieved by using varying additions of a 10 % (by volume) solution of CH_3NO_2 in ethanol as an imposed quenching agent. The [54]Mn cocktails were in all cases composition matched to the [103]Pd cocktails by the addition of proportionate amounts of the blank carrier solutions that were used to prepare the master solutions.

In the second case, the same master solutions ML and MH were assayed for [103]Pd activity by the Triple-to-Double Coincidence Ratio (TDCR) method, which is a quasi-absolute LS technique that uses a custom-made three-phototube spectrometer to determine the detection efficiency internally without the need for efficiency tracing standards. The spectrometer was constructed at NIST using a similar system in operation at LNHB (see Zimmerman, *et al.* [10] and references therein). The technique has become widely accepted for use with β-emitting and some EC radionuclides, although the technique is less established in the case of EC. For these measurements, a total of six series of cocktails, each with three independently prepared cocktail samples, were prepared using three different commercially available scintillants. The detection efficiency was varied for each cocktail by changing the voltage on the focusing electrodes of the three phototubes. Each cocktail was counted three times for 5 minutes livetime at each of 9 focusing electrode voltage settings. For most of the cocktail samples, this procedure was repeated an additional time approximately one week later. Detection efficiencies for the double coincidence counting channel were calculated as a function of the TDCR using the program *Pd103_2* [7], assuming equal detection efficiencies for each phototube. The program was modified to ensure consistency between the program's input nuclear and atomic decay data with the data in reference [1].

A large number of components of the uncertainty on the activity measurements were identified and evaluated. These included components from the dissolution and gravimetric dilution scheme, gravimetric determinations of LS sample aliquants, LS measurement precision (for both within- and between-measurement-occasion variability), nuclear decay data (for both [103]Pd and [54]Mn) and detection efficiency calculations, LS spectrometer livetime determinations, [54]Mn standard activity, and decay corrections. The expanded uncertainties on the [103]Pd activity determinations were 1.5 % and 2.3 % for the TDCR and LSET techniques, respectively.

Table 12 summarizes the results for the comparison of the three primary standardizations methods. As indicated, the calorimetric result agrees with the LSET values to about 2 % and with the TDCR results to about 4 %. The relative expanded ($k = 2$) uncertainties in the massic activities for the master solutions are approximately ± 2 % for all three methods. The results, therefore, can be considered to be in agreement to within the measurement uncertainties and can be considered to have confirmed the validity of the calorimetry to within its measurement uncertainty. Nevertheless, there is the appearance of a positive bias in the LS spectrometry compared to the calorimetry. Furthermore, it is known that the TDCR values may contain a possible systematic error (of a still undetermined magnitude and for which no corrections were made) that resulted from known asymmetric phototube responses in the TDCR system.

Table 12. Comparison of the two LS-based standardizations to the calorimetric results in terms of ^{103}Pd massic activity for the master solutions MH and ML at a Reference Time of 1200 EST 6 May 2002.

Standardization method	^{103}Pd massic activity (MBq g^{-1})		
	Solution MH	Solution ML	MH/ML ratio
Calorimetry	91.46	1.612	56.74
LSET	93.68	1.638 *	57.19 *
TDCR	95.54	1.672 *	57.16 *

* These results may be influenced by solution inhomogeneity.

For completeness, it can be noted that the calorimetric results cited here and throughout this Report are based on use of an **assumed** average energy per decay of (60.82 ± 0.08) keV, which corresponds to (9.744 ± 0.013) µW·GBq^{-1}. It is entirely possible that this value could change by as much as ± 1 % to ± 2 % with improved knowledge of the ^{103}Pd decay scheme data, with corresponding changes in the calorimetric results reported here. Any changes in the decay data would affect the LSET and TDCR efficiency calculations as well. However, the calorimetry is likely to be much less sensitive to changes in the ^{103}Pd decay scheme data than the LS results.

XII. Photonic-emission spectrometry

High-resolution photonic–emission spectrometry was performed using four different photon detection systems (viz., the NIST "B", "T", "X", and "S" detectors) on point sources and on 5-mL ampoules that were prepared from aliquants of the master solutions MH and ML, as well as on the sources L3 and H1 directly.

Examinations for radionuclidic impurities using source L3 at 45 cm with the "T detector" [a p-type closed-end high-purity germanium (HPGe) coaxial detector with an active crystal volume of about 80 cm^3 which has an inactive germanium region of about 0.3 µm and a 0.5-mm thick aluminum-absorbing layer] resulted in identifying traces of ^{65}Zn and ^{105}Ag, both with activities less than 0.001 % of the ^{103}Pd activity at the 1200 EST 6 May 2002 reference time.

Activity measurements on an ampoule source (denoted MH8) that was prepared from the MH master solution with both the "X and "T" detectors, as derived from photon emission rates, resulted in values ranging from 88 MBq·g^{-1} to 122 MBq·g^{-1}. The inability to achieve consistency in any of the activity determinations by photonic-emission spectrometry using the currently available data for the photon emission probabilities underscores problems with the ^{103}Pd decay scheme and the need to improve this data base. Although the dubious accuracy of the nuclear and atomic data used to calculate the activity could be identified as one primary cause for these discrepancies, the influence of efficiency determinations for the detectors at low energy, as well as the correct application of appropriate correction factors (for coincident summing and escape phenomena), also requires further investigation. Until these issues are resolved, it will be impossible to reconcile the calorimetric and LS activity standardizations with activities derived from photon emission measurements.

III. Cited references

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