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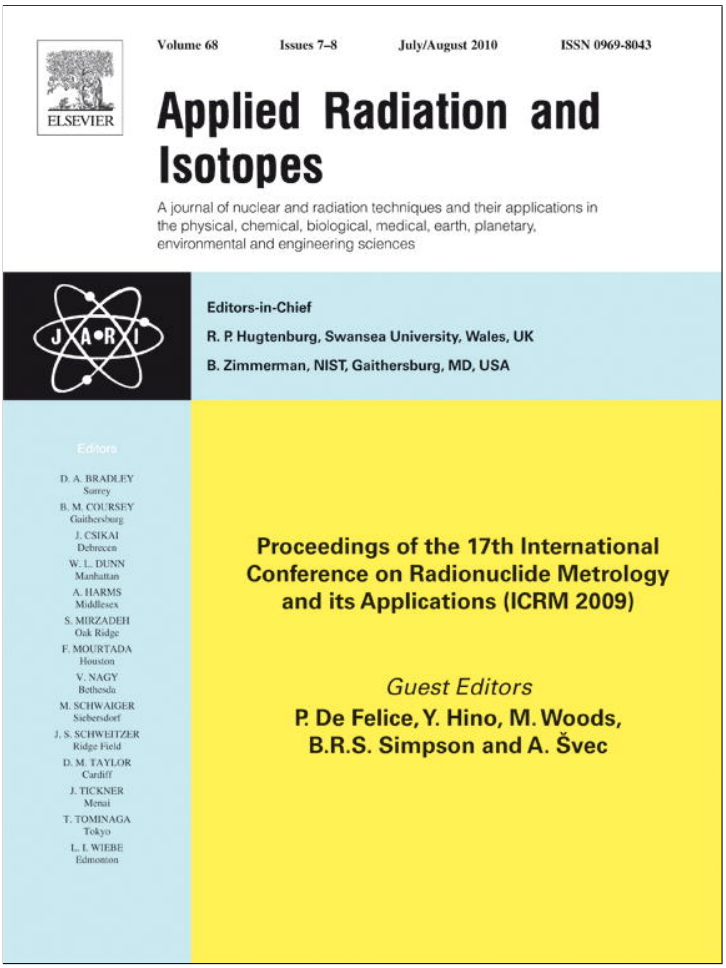

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Investigation into the standardization of ^{99}Tc

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ABSTRACT

The standardization of ^{99}Tc by several primary methods was investigated. This was performed to support a new ^{99}Tc transfer standard that has been developed and will be disseminated by the National Institute of Standards and Technology (NIST) as Standard Reference Material SRM 4288B. The standardization for the ^{99}Tc content of the solution was based on $4\pi\beta$ liquid scintillation (LS) measurements with ^{3}H-standard efficiency tracing (CIEMAT/NIST method). Confirmatory determinations were performed by $4\pi\beta$(LS)-γ(NaI) live-timed anti-coincidence (LTAC) counting and an LS-based $4\pi\beta$ triple-to-double coincidence ratio (TDCR) method.

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1. Introduction

This paper describes the development of a ^{99}Tc solution standard at the National Institute of Standards and Technology (NIST). Technetium-99 is a pure beta emitter with a half-life of $(2.111 \pm 0.012)\,10^5$ y (Peker, 1994). It is the most significant long-lived fission product and one of the more significant components of nuclear waste. It can be obtained by irradiating molybdenum with deuterons. A ^{99}Tc standard is important for monitoring inside nuclear facilities and in the environment. Since there is no stable technetium isotope, and ^{99}Tc is the longest lived of its isotopes, it is also used in chemical studies to characterize the radiopharmaceuticals developed for ^{99m}Tc. In addition, most of the chemistry for the element is based on studies with ^{99}Tc.

Technetium-99 is a beta emitter (Peker, 1994) with a decay probability to ground state of $I_\beta = 0.999984 \pm 0.000004$ [E_β(max)$=(293.5 \pm 1.4)$ keV; E_β(ave)$=(84.6 \pm 0.5)$ keV]. This is a second forbidden non-unique transition ($9/2^+$ to $5/2^+$; $\Delta J=2$, $\Delta\Pi=0$). It also decays to the short-lived (18.9 ± 1.0) ns, 89.50 keV, isomeric state with a probability of $I_\beta = 0.000016 \pm 0.000004$ (Peker, 1994). However, only a negligible part (0.000065%) of the ^{99}Tc decay occurs through the 89.5 keV γ-ray.

A previous solution of ^{99}Tc (SRM 4288A) was calibrated in 1996 by $4\pi\beta$(LS)-γ(NaI) live-timed anti-coincidence (LTAC) counting with ^{60}Co efficiency tracing and by $4\pi\beta$ liquid scintillation (LS) spectrometry (Lucas, 1998). The LS calibration showed dependence on the ^{99}Tc beta spectrum shape factor. Analysis was performed assuming allowed, 2nd forbidden and experimental "partially forbidden" transitions using the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) developed EFFY code (Grau Malonda and Garcia-Toraño, 1982; Coursey et al., 1985, 1986). The same analysis is performed here using the TRACER code developed by Cassette (2006).

The beta transition class is an important factor in the calculation of the efficiency as determined by the various codes available (i.e., TRACER code (Cassette, 2006), CN2003 (Gunther, 2003) and EFFY4 (Garcia-Toraño, 1993)) to use in the efficiency tracing method. The outputs of these codes are based on data that includes, among others, the transition class, the end point and/or average energy, the beta spectrum shape, an ionization quenching function, and scintillant. In order to simplify the calculations, certain assumptions are made that need to be taken into account when performing any analysis (Laureano-Pérez et al., 2007).

The shape factor for a second forbidden non-unique transition is given by $C(W)=q^2+p^2$, where p is the electron momentum and q is the neutrino momentum. However, an experimental shape factor for ^{99}Tc was found to be $C(W)=q^2+(0.54 \pm 0.02)p^2$ (Reich and Schüpferling, 1974). This is an experimental "partially forbidden" transition with a value between a second forbidden non-unique transition and an allowed transition $C(W)=1$. The beta efficiencies used in CNET[2] were affected by these shape factors (Lucas, 1998).

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[1] The National Institute of Standard and Technology is an agency of the US Department of Commerce.

[2] The acronym CNET refers to CIEMAT/NIST Efficiency Tracing method. CIEMAT/NIST refers to the two laboratories that collaborated in developing the protocol for this LS tracing methodology; viz., the Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas (CIEMAT) and the National Institute of Standards and Technology (NIST).

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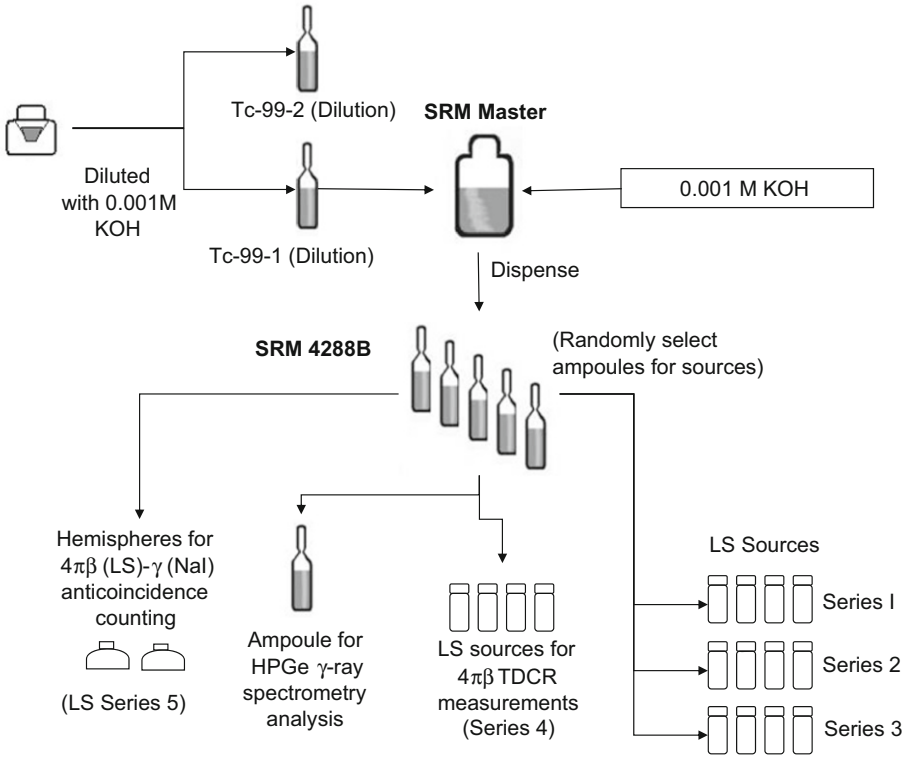

Fig. 1. Scheme for preparation of the ^{99}Tc solution standards (SRM 4288B) and the counting sources used for the $4\pi\beta$ LS-based standardization, impurity determination and confirmatory measurements.

2. Experimental methods, results and discussion

2.1. Overview

Fig. 1 illustrates the scheme used to prepare the ^{99}Tc standard and counting sources used in the LS analysis and confirmatory measurements performed at NIST. The certified ^{99}Tc massic activity for the standards was obtained from a primary standardization based on CNET $4\pi\beta$ liquid scintillation (LS) spectrometry. Confirmatory measurements were performed by: (i) $4\pi\beta$(LS)-γ(NaI) live-timed anti-coincidence (LTAC) counting; and (ii) an LS-based $4\pi\beta$ triple-to-double coincidence ratio (TDCR) method.

2.2. Preparation of master solution and SRM ampoules

The master solution was prepared carrier-free from a stock solution obtained from Isotope Products Laboratory (IPL)[3] that was diluted with a 0.001 mol L^{-1} KOH solution. The master solution density was determined to be (0.997 ± 0.002) g mL^{-1} at 22.5 °C. Nominal 5 mL aliquants of the master solution were dispensed into 5 mL borosilicate-glass ampoules. The average solution mass per ampoule, as determined from the dispensed mass differences, was (4.9945 ± 0.0003) g and was based on measurements of 12 ampoules (about every 10 apart in sequence) from a set of 100 ampoules. After filling, the SRM ampoules were flame-sealed and sterilized by autoclaving at about 200 kPa for 20 min.

[3] Certain commercial equipment, instruments and materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials and/or equipment are the best available for the purpose.

2.3. Photonic-emission impurity analyses

Analysis for photon-emitting impurities was performed using a high purity intrinsic germanium detector: B-detector (L=54 mm; φ=55 mm). The detector is an n-type coaxial detector with a 0.5 mm Be window. The detector and spectrometry procedures used by NIST have been described by Pibida et al. (2006, 2007).

No impurity photons were detected. A lower limit of detection, expressed as a massic photon emission rate, on 29 February 2008, was: 1.2 s^{-1} g^{-1} for energies between 30 and 3600 keV. The relative standard uncertainty contribution to the certified ^{99}Tc massic activity due to photon-emitting impurities was estimated to be equal to the estimated limit of detection for the impurities.

2.4. LS-based primary standardization

2.4.1. CNET methodology

The certified massic activity for ^{99}Tc was obtained by $4\pi\beta$ liquid scintillation (LS) spectrometry using the CNET method. The LS detection efficiency was calculated using the TRACER code (Cassette, 2006) for CNET with composition-matched LS cocktails of a ^{3}H standard as the efficiency detection monitor. The ^{3}H nuclear data used in the calculation was obtained from Chechev (2006). This method has been described in detail elsewhere along with the specific analysis procedure used for this work (Collé and Zimmerman, 1997; and references therein).

2.4.2. LS cocktails

Due to the alkaline nature of the ^{99}Tc solution, the pseudocumene-based scintillant, Hionic Fluor (HF) from Perkin Elmer, Waltham, MA was used. Five series of cocktails were prepared and the compositions are summarized in Table 1. The cocktails were

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Table 1
The Hionic Fluor (HF) LS cocktail compositions used by NIST for standardization of ^{99}Tc.

Cocktail series	Scintillant	N_s	f_w	Quenching agent	Aliquat® 336
1	HF	4	0.04–0.17	H_2O	No
2	HF	6	0.03	CCl_4	Yes
3	HF	6	0.03	CCl_4	No
4	HF	3	0.03–0.06	N/A	Yes/No
5	HF	6	0.02–0.06	N/A	Yes

N_s=number of samples in each series; f_w=H_2O mass fraction in cocktail.

prepared from randomly selected ampoules in the set of SRM 4288B ampoules.

Since the contact of nitromethane, a routinely used quenching agent, with KOH could cause the formation of shock-sensitive salts a different quenching agent was needed. Based on observations during a ^{209}Po standardization (Collé et al., 1995), water was chosen as a first approach. With water as the quenching agent ranging in aqueous fraction from 0.04 to 0.17, the ^{3}H efficiency was found to vary by only about 10% to 12%. Water, obviously, was not a good quenching agent since large amounts of it would be needed to obtain a wide range of efficiencies. A second experiment was performed comparing the effect of carbon tetrachloride (CCl_4) as opposed to water on scintillant quenching based on changes in the quenching parameters on three different LS counters. Approximately 0.5 mL of water and CCl_4 was added to two identical cocktails. The quenching parameters were measured and compared to a blank. Carbon tetrachloride caused 62 times more change in the quenching indicating parameter Horrocks Number (H#) than water. Series 2 and 3 then used CCl_4 as a quenching agent.

In the 1996 LS sources, a mixture of quaternary ammonium salts (Aliquat® 336) was used as a surfactant and/ or complexing agent for the pertechnetate ion in the cocktail preparation (Lucas, 1998). In the present 2008 cocktails, two series of LS cocktails were prepared: one with Aliquat® 336 to reproduce the composition of the 1996 counting source (series 2); and one without (series 3). The massic activity value between these two sets differed by 0.05% demonstrating the negligible effect of the use of this surfactant in addition to the amount of surfactant inherent to the scintillant. Aliquat® 336 was added to series 2, 4 and 5. Series 1–4 were contained in 20 mL glass vials fitted with polyethylene lined plastic caps. Series 1–3 were used for the CNET measurements while series 4 was used in the LS-based TDCR measurements. In series 4, two of the samples were prepared without Aliquat® 336 and a water fraction of 0.06. The other sample was prepared with Aliquat® 336 and a 0.03 water fraction. The series 5 samples were contained in specially fabricated, glass pseudo-hemispheres for LTAC counting (See Section 2.5.1).

In series 1–3, an equal number of composition-matched ^{3}H cocktails were prepared using a gravimetric dilution of NIST tritiated-water standard SRM 4927F (NIST, 2000). These ^{3}H LS sources were matched to the ^{99}Tc sources in terms of composition by the addition of non-radioactive carrier solutions. The sources consisted of an appropriate aliquant of either ^{99}Tc or ^{3}H solution, approximately 9.5 mL of Hionic Fluor, and an aqueous fraction range of 0.04–0.17 for series 1 and 0.03 for series 2 and 3. Varying masses of an imposed chemical quenching agent, carbon tetrachloride, were added to the ^{3}H and ^{99}Tc cocktails within a given series (2 and 3) to vary the LS detection efficiencies over a wide range. Blank sources for background subtraction were composed of scintillant, carrier solution (to match compositions), and carbon tetrachloride (to match chemical quenching). All of the mass additions of radioactive aliquants (^{99}Tc and ^{3}H), typically 31–70 mg, were performed with a microbalance using polyethylene aspiration-type pycnometers.

2.4.3. LS spectrometers

Three different instruments were used for the LS measurements: (i) Packard TriCarb A2500 TR (Perkin-Elmer, Wesley, MA); (ii) Wallac 1414 Winspectral (Perkin-Elmer, Wesley, MA) and (iii) Beckman LS 6500 (Beckman Coulter, Fullerton, CA). They have considerably different operating conditions characteristics (e.g., low-energy threshold, deadtime, linear vs. logarithmic energy binning, etc.), which helps to ensure variation in measurement conditions during standardization. A more detailed description is presented elsewhere (Laureano-Pérez et al., 2007).

2.4.4. $4\pi\beta$ LS-based standardization result and uncertainty

Three distinct sets of LS measurements were performed, corresponding to the three sets of cocktails. The counting arrangement used for each set consisted of interspersing the matched pairs of ^{99}Tc and ^{3}H cocktails into the counters, with at least two blanks positioned at certain intervals within the series. The counting time was typically 20 min per sample for three to five cycles per measurement occasion. Each cocktail series was counted on all three counters. In addition, they were measured on more than one occasion in at least one counter. The measurements for a given series were conducted over periods ranging from about 16 to 28 days. Table 2 shows the results for the 192 determinations of the ^{99}Tc massic activity (at a 1200 EST 1 May 2008 reference time) as derived from measurements obtained on 11 separate occasions over the course of 28 days with the series 1–3 cocktails. Due to the small efficiency range obtained during CNET analysis on series 1, the results from this series were not considered to be sufficiently reliable. Therefore, the standardization was based exclusively on the results from series 2 and 3. For these two series, the massic activity results were in very good agreement within each series (i.e. between instruments within series), with a relative standard deviation of the mean of 0.06%. The difference between averages for the two series was 0.05%. The unweighted mean average of series 2 and 3 was used as the certified value for the ^{99}Tc massic activity, $A=(31.55 \pm 0.21)\,\mathrm{kBq\,g^{-1}}$ for k=2 at 1200 EST 1 May 2008. A normality test on all the series was performed (Dataplot, 2007) and all results could accept the normality assumption at a 95% and 99% level.

Table 2
Results for the massic activity for SRM 4288B, at a reference time of 1200 EST 1 May 2008, as obtained from the CNET measurements at NIST.

Series	Instrument	Age/ d	N_d	$\varepsilon_{H\text{-}3}$	$\varepsilon_{Tc\text{-}99}$	A (kBq g^{-1})	s (%)
1	B	1	12	0.239-0.263	0.927–0.933	32.41	0.029
	P	4	12	0.194-0.216	0.916–0.922	32.42	0.008
	W	28	12	0.193-0.217	0.916–0.922	32.47	0.007
2	B	2	18	0.274-0.398	0.935–0.954	31.55	0.031
	P	6	30	0.209-0.317	0.920–0.942	31.52	0.027
	W	12	18	0.208-0.321	0.920–0.943	31.56	0.026
	B	16	18	0.270-0.395	0.934–0.953	31.56	0.056
3	P	2	18	0.194-0.326	0.916–0.944	31.54	0.035
	B	6	18	0.248-0.397	0.930–0.953	31.57	0.014
	W	12	18	0.194–0.326	0.916–0.944	31.59	0.026
	P	16	18	0.192–0.324	0.916–0.943	31.55	0.034
2 & 3	Unweighted grand mean					**31.55**	–
2 & 3	Relative standard deviation of mean (n=8)					–	**0.02**

Refer to Table 1 for LS cocktail compositions of the series. Instrument refers to LS counter used: W=Wallac; B=Beckman; P=Packard. Age is the approximate time between LS cocktail preparation and the middle of the counting cycles in days; and N_d is the number of determinations. The $\varepsilon_{H\text{-}3}$, $\varepsilon_{Tc\text{-}99}$ are the range of values for the ^{3}H and ^{99}Tc LS efficiency in the series of the quenched cocktails, respectively. A is the massic activity in units of kBq g^{-1} at reference time 1200 EST 1 May 2008 and s is the relative standard deviation (for N_d determinations) in percent.

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This ^{99}Tc solution was intercompared to the previous ^{99}Tc standard (SRM 4288A) calibrated at NIST by Lucas (1998). The two massic activity measurements were in agreement to within −0.5%. The relative k=2 expanded uncertainties on the determination of SRM 4288A is approximately 1.1%.

The relative combined standard uncertainty of the ^{99}Tc massic activity is 0.33%. The relative expanded uncertainty, using a coverage factor k=2, is 0.66%, which is assumed to provide an approximate 95% level of confidence. Table 3, as taken directly from the SRM 4288B certificate (NIST, 2008), shows the component uncertainties used to obtain these estimates. The major contributor to the overall uncertainty was the model dependence on the efficiency determination. This can be easily explained by the effect of the beta spectrum shape factor on the efficiency calculated for a double tube system. Fig. 2 shows the effect of using a second forbidden non-unique shape factor of $C(W)=Aq^2+B\lambda_2 p^2$ (Behrens and Szybisz, 1976) where $A=B=1$ and with variable λ_2. A change in the λ_2 parameter from 0.54 to 1 results in a 0.14% to 0.3% uncertainty, depending on the ^{3}H efficiency range (even though ^{99}Tc has a high efficiency which makes its activity determination less dependent on efficiency). The uncertainty on the value of experimental λ_2 parameter results in an uncertainty on the ^{99}Tc efficiency of up to 0.18%. The value of the Birks parameter (kB) was also varied in order to assess its effect on the efficiency obtained. No significant effect was observed in the efficiency by its variation.

2.5. Confirmatory measurements

The intent of any confirmatory measurement as performed by our laboratory is to ensure that the standardization result is

Table 3
Standard uncertainty components for the ^{99}Tc massic activity of SRM 4288B, calibrated by $4\pi\beta$ LS spectrometry with ^{3}H-standard efficiency tracing.

	Uncertainty component	Assessment type[a]	Relative standard uncertainty contribution on massic activity of ^{99}Tc (%)
1	LS measurement repeatability; reproducibility in massic activity for 2 different cocktail compositions, with 6 samples in each, measured in 3 counters on 1 or 2 measurement occasions; standard deviation of the mean for n=8 data sets, normally distributed. The LS within-measurement precision for a given data set, in terms of the standard deviation of the mean for 6 samples measured for 3 to 5 cycles on three measurement occasions, ranged from 0.09% to 0.12%	A	0.06
2	Background; wholly embodied in 1	A	–
3	LS counters dependencies wholly embodied in components 1 & 2	A	–
4	Gravimetric (mass) measurements for preparation of sources	B	0.1
5	Live time determinations for LS counting time intervals, includes uncorrected dead time effects	B	0.06
6	^{3}H decay corrections for half life uncertainty of 0.16%	B	0.002
7	^{99}Tc decay corrections for half-life uncertainty of 0.57%	B	3×10^{-7}
8	Limit for photon-emitting impurities	B	0.004
9	Computed β detection efficiencies (model dependencies and computed β spectra)	B	0.3
	Relative combined standard uncertainty		**0.33**
	Relative expanded uncertainty (k=2)		**0.66**

[a] (A) denotes evaluation by statistical methods; (B) denotes evaluation by other methods.

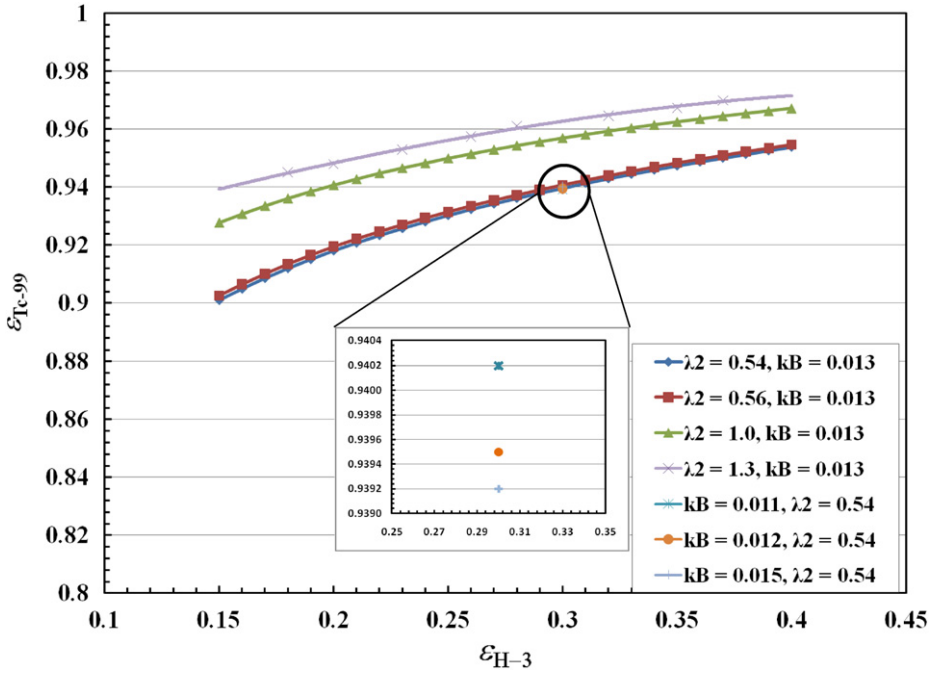

Fig. 2. Comparison of the computed LS efficiency for the ^{99}Tc β transitions in a double tube coincidence system (ε_{Tc-99}) as obtained with the *TRACER* code using various shape factors (λ_2) values. All were calculated with kB=0.013 cm MeV^{-1}. The Birks kB parameter was also varied. Additional points with kB=0.011, 0.012 and 0.015 cm MeV^{-1} were calculated at a ^{3}H efficiency (ε_{H-3})=0.3 with λ_2=0.54.

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confirmed by supplementary measurements that are completely independent of the primary method. Confirmatory measurements were performed by: (i) $4\pi\beta$(LS)-γ(NaI) live-timed anti-coincidence (LTAC) counting; and (ii) an LS-based $4\pi\beta$ triple-to-double coincidence ratio (TDCR) method. The certified value was in agreement with the confirmatory determinations to within −0.22% and +0.06%, respectively, for the LTAC and TDCR methods. The relative k=2 expanded uncertainties on the two confirmatory determinations are approximately 0.7% and 0.3%, respectively.

2.5.1. $4\pi\beta$(LS)-γ(NaI) anti-coincidence counting

A confirmatory determination of the ^{99}Tc activity for the SRM solution was performed by live-timed $4\pi\beta$(LS)-γ(NaI) anti-coincidence counting with ^{60}Co efficiency tracing (series 5). Details on the earlier application of the ^{60}Co efficiency tracing method by NIST to ^{99}Tc were presented by Lucas (1998). For the present work, 5 LS hemisphere sources were prepared containing 4 different ^{99}Tc to ^{60}Co ratios (8.0, 2.8, 0.94, 0.55). Also, two sources containing no ^{99}Tc were prepared. Each source contained 4-mL of Hionic Fluor LS cocktail, 1 drop of Aliquat® 336 and technetium and cobalt carrier to match the composition among the various activity ratios. The total aqueous fraction was about 0.03. The β–γ anti-coincidence data from each mixed-nuclide source was fit with a linear function over an efficiency domain of 0.55–0.93 and with a quadratic fit over a range of 0.40–0.93. The ^{60}Co-only sources were analyzed in the same manner and the ^{60}Co activity was subtracted from intercepts of the mixed sources. The standard deviation of the massic activities determined from the 4 mixed sources was 0.15% and 0.18% for linear and quadratic fits, respectively. The difference between the average activity determined by linear and quadratic fits was 0.29%. There was no discernible trend in the residuals from either fit. Therefore, the final massic activity was determined by averaging the linear and quadratic fit results. The fitting uncertainty was the dominant uncertainty in the result.

2.5.2. LS-based $4\pi\beta$ triple-to-double coincidence ratio (TDCR) method

The TDCR measurements were carried out between 1 May 2008 and 8 August 2008 using the LS cocktails prepared as described above. Specifically, three radioactive sources and two background blanks, which had identical cocktail compositions (in terms of aqueous fraction, ion content, etc.), were prepared for the TDCR measurements (series 4).

Measurements were made using the NIST TDCR spectrometer, which has been previously described (Zimmerman et al., 2004a, b). Each source, including the background blanks, was measured on three occasions during the time period noted above. Each counting experiment consisted of four repeated measurements at each of eight efficiency points, which were obtained from the use of four grey filters used at each of two focusing electrode voltages.

The counting data were analyzed using a program developed in-house for use with the *Mathematica* (Wolfram Research, 2001) symbolic mathematics package. The input data consist of the beta spectrum for ^{99}Tc, and the value of the Birks kB parameter, which was taken to be 0.012 cm MeV^{-1}. The beta spectrum was calculated using the program SPEBETA (Cassette, 1992) using the endpoint energy given above and the shape function for a second forbidden non-unique decay, which is $C(W)=q^2+\lambda_2 p^2$, with an experimental value of $\lambda_2=0.54\pm0.02$ for ^{99}Tc (Reich and Schüpferling, 1974).

The stopping power, dE/dx, for electrons in the LS cocktail was calculated by fitting a function of the form dE/d$x=a+bE+c(\ln E)^2+d\ln E/E+e/E$ (E is the value of the midpoint energy for each bin of the calculated beta spectrum) to data from the NIST ESTAR (NIST, 2007) database using the compositions given in (LNHB, 2007) for Ultima Gold. The differences in scintillation composition contributed 0.0036% (k=2) to the combined standard uncertainty.

The program calculates the individual phototube efficiencies, thereby allowing for correction due to asymmetry in the counting rates in each of the doubles counting channels. The efficiencies for the "logical sum of doubles" channels, as calculated from the program, ranged from 0.81 to 0.92.

A total of 72 determinations of the massic activity were used in the calculation of the central value and consisted of the average of the four repeated measurements taken at eight efficiencies for each of the three sources on three different occasions. The central value was taken as the average of these results, and was found to be (31.53 ± 0.04) kBq g^{-1} as of 1200 EST 1 May 2008, where the uncertainty is a standard uncertainty. The deviation in measurement repeatability was the dominant uncertainty in the result.

3. Summary

The ^{99}Tc radioactivity solution standards were certified and will be disseminated as NIST SRM 4288B with the following specifications: *Solution mass*: (4.9945 ± 0.0003) g; *Solution density*: (0.997 ± 0.002) g mL^{-1} at 22.5 °C; *Massic activity*: 31.55 kBq g^{-1}; *Relative expanded* (k=2) *uncertainty*: 0.66%; *Reference time*: 1200 EST, 1 May 2008.

The certified massic activity for ^{99}Tc was obtained by $4\pi\beta$ liquid scintillation (LS) spectrometry using the CNET method. Analysis was performed assuming allowed, 2nd forbidden and experimental "partially forbidden" transitions. The uncertainty on the shape factor causes an uncertainty on the efficiency of up to 0.3% based on the ^{3}H efficiency range.

Confirmatory measurements were also performed by: (i) LTAC counting; and (ii) TDCR methods. The certified value was in agreement with the confirmatory determinations to within −0.22% and +0.06%, respectively, for the LTAC and TDCR methods. The relative k=2 expanded uncertainties on the two confirmatory determinations are approximately 0.7% and 0.3%, respectively. Massic activity measurements from an intercomparison of this ^{99}Tc standard to a previously certified ^{99}Tc standard agreed within −0.5%.

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Discussion

Due to technical problems (unclear records) the transcription can not be published.