

ACTIVITY STANDARDIZATION of ^{99m}Tc USING LIQUID SCINTILLATION COUNTING METHOD

K. B. Lee, Jong Man Lee, Tae Soon Park, Sang Han Lee

Korea Research Institute of Standards and Science, Daejeon 305–600, Korea

The Korea Research Institute of Standards and Science (KRISS) participated in the international comparison of activity measurements of ^{99m}Tc that was held at KRISS in 2010. We develop the national primary standard for ^{99m}Tc solution used during the comparison. We use two primary liquid scintillation counting techniques for the standardization: digital coincidence counting with the 4πβ(LS)-γ(NaI(Tl)) system and triple-to-double coincidence ratio counting. Both methods give consistent results for the specific activity of a ^{99m}Tc solution. Adopting the result from the digital coincidence counting method as the reference value of the ^{99m}Tc measurement, we evaluate the standard uncertainty of 0.856% for the KRISS primary standard of the ^{99m}Tc activity. The comparison result shows that the newly-established KRISS primary standard lies within the standard uncertainty with the Key Comparison Reference Value defined from other ^{99m}Tc measurements.

I. INTRODUCTION

The metrological comparability of Technetium-99m measurements is hard to establish because of the low energy of its emitted radiation in coincidence with gamma rays as well as its short half-life. The short half-life of ^{99m}Tc prevented the Korea Research Institute of Standards and Science (KRISS) from sending a ^{99m}Tc ampule for measurement at the Bureau International des Poids et Mesures (BIPM). KRISS participated in the BIPM comparison of activity measurements of ^{99m}Tc that was held at the KRISS in September 2010. This paper describes the national primary standard for ^{99m}Tc activity calibrating the reference ionization chamber that actually measured the ^{99m}Tc solution used during the comparison.

II. EXPERIMENTAL

We measure the activity of a ^{99m}Tc solution using two liquid scintillation (LS) counting methods: Digital Coincidence Counting (DCC) with the 4πβ(LS)-γ(NaI(Tl)) system [1][2] and Triple-to-Double Coincidence Ratio (TDCR) counting [3]. Two kinds of LS cocktails are used to investigate the dependence of our measured activity on the LS solvent used: pseudocumene-based Hionic Fluor and DIN-based Ultima Gold.

II.A. Digital coincidence counting with the 4πβ(LS)-γ system

The 4πβ(LS)-γ system operated in DCC mode acquires the pulse-height spectra for the LS and NaI channels for each LS source prepared. The β-γ coincidence counts are recorded using the arrival time differences between the two sequences of detector pulses: the pulses from the liquid scintillation detector and the pulses from the NaI(Tl) detectors resulting from the 141 keV gamma-ray energy emanating from the decays of the ^{99m}Tc radionuclides. The eight active sources are each counted for 10 min and the two blank sources for 20 h. For one typical source, Figures 1 (a) and (b) present the pulse-height distributions of the two LS channels, Figure 1 (c) shows the distribution of the sum of coincident pulses of the two LS channels, and Figure 1 (d) displays the pulse-height distribution of the gamma channel. The 141 keV gamma-ray peak in Figure 1 (d) is evident. The single-electron peak in Figure 1 (c) is greatly reduced in comparison with those of Figure 1 (a) and (b) owing to the coincidence requirement of the two LS channels. Present in Figure 1 (c) are the two broad peaks in addition to the single-electron peak. The two broad peaks are expected to be due to the about 20 keV Auger electrons and about 120 keV conversion electrons from ^{99m}Tc decays. Since the 120 keV conversion electrons emanate from the gamma transitions from the 141 keV energy level to the ground level of ^{99m}Tc, they are not in coincidence with the 141 keV gamma-rays.

For each threshold value various rate-dependent systematic effects such as dead time, coincidence resolving time and out-of-channel effects are corrected to obtain the true beta rate ρ_β , the true gamma rate ρ_γ and the true coincidence rate $\rho_{\beta\gamma}$.

The data points to be extrapolated for activity determination are the experimental values of $\rho_{\beta}\rho_{\gamma}/\rho_{\beta\gamma}$ as a function of $\rho_{\gamma}/\rho_{\beta\gamma}-1$ at various discrimination levels. Figure 2 shows $\rho_{\beta}\rho_{\gamma}/\rho_{\beta\gamma}$ as a function of $\rho_{\gamma}/\rho_{\beta\gamma}-1$ for one ^{99m}Tc source and a superimposed linear fit obtained using a generalized least-squares method. The measured activity is obtained from the y-axis intercept of the fitted line, which turns out to be (11.90 ± 0.19) kBq. The quoted uncertainty is the standard uncertainty evaluated by the chi-square method [4] representing the repeatability of the result. The specific activity is derived using the mass of transferred active solution for the source. This method gives a reported activity value with the relative standard uncertainty of 0.856%, the dominant component of which is due to the reproducibility with the fit ranges used.

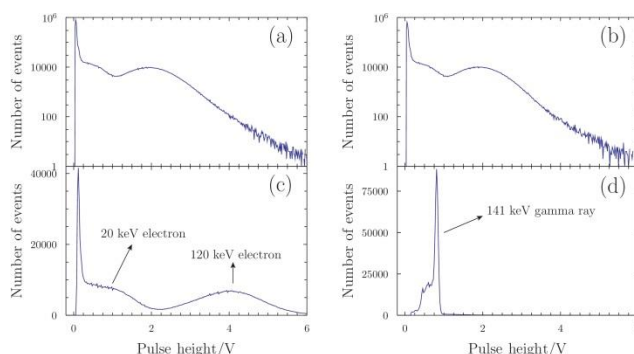


Figure 1: Pulse-height distributions for the ^{99m}Tc pulses. (a) and (b) Two dynode signals of the beta channels. (c) Sum of the two coincident dynode signals of the beta channels. (d) Sum of the two gamma channels

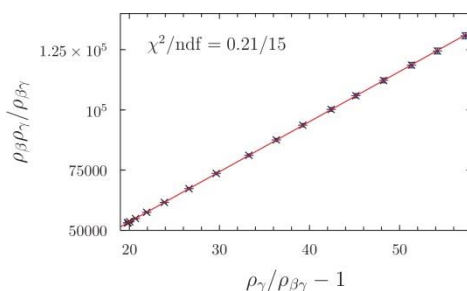


Figure 2 Efficiency extrapolation for ^{99m}Tc

II.B. Triple-to-double coincidence ratio counting

The same LS sources are counted using the KRISS TDCR system. We calculate the double-coincidence efficiency and triple-coincidence efficiency for ^{99m}Tc as a function of the TDCR parameter. The calculation is based on the Poisson distribution for the number of photoelectrons in the PM tubes. The ionization quenching is corrected by using the Birks formula with Birks kB as a free parameter. The stopping power of electrons in the LS cocktail, which is necessary for the Birks formula, is calculated from the formula for the linear energy transfer of electrons. Figure 3 shows the calculated double-coincidence and triple-coincidence efficiencies of ^{99m}Tc as a function of the calculated TDCR parameter. The calculation is based on the simulation of the Ultima Gold cocktail with kB value equal to $0.0025 \text{ cm}\cdot\text{MeV}^{-1}$. From the figure we see that the calculated double-efficiency value corresponding to the mean experimental TDCR parameter of 0.79 is 24%. The mean of the measured specific activities of the four Ultima Gold sources, along with its standard deviation, is 18.491 (94) MBq/g. The same procedure applied to the Hionic Fluor sources gives a mean specific activity 18.390 (80) MBq/g. However, in this case the calculation is performed at a different kB value of $0.007 \text{ cm}\cdot\text{MeV}^{-1}$.

III. CONCLUSIONS

The compatibility of the measured activity using the DCC method is established by comparisons of the result with those obtained with the TDCR method and the ionization chamber method. In the TDCR method the double-coincidence efficiency and triple-coincidence efficiency are calculated on the basis of the Poisson distribution for the number of photoelectrons in the photomultiplier (PM) tubes. The K4 comparison result [5], as displayed in Figure 4, shows that the KRISS primary standard for ^{99m}Tc activity lies within the standard uncertainty with the Key Comparison Reference Value, defined from other ^{99m}Tc measurements.

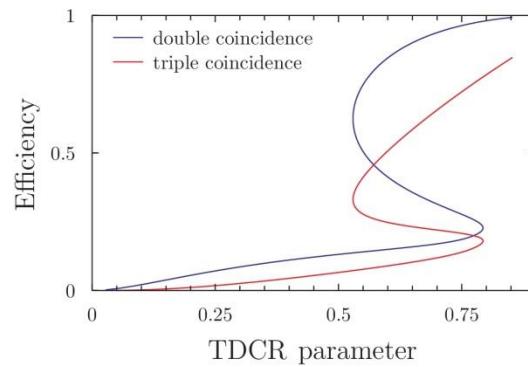


Figure 3 Calculated double-coincidence and triple-coincidence efficiencies of ^{99m}Tc function of the calculated TDCR parameter.

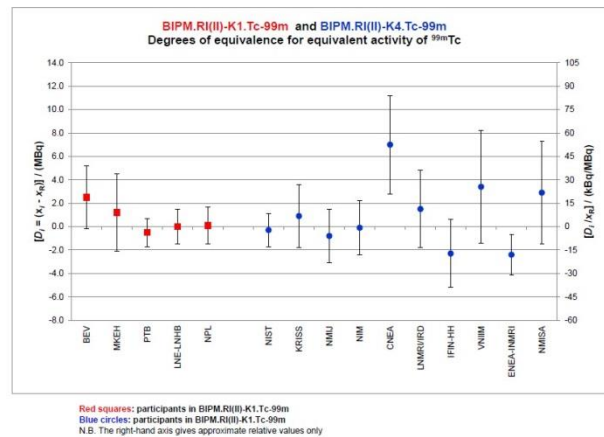


Figure 4 International comparison of activity measurements of ^{99m}Tc

REFERENCES

1. Lee, K. B., Lee, J. M., Park, T. S., Lee, S.H., oct. 2010b. Development of a high-efficiency $4\pi\beta(\text{LS})-\gamma$ coincidence system for direct measurements of activity in radioactive decay. Nuclear Science, IEEE Transactions on 57 (5), 2613-2616.
2. Lee, K. B., Lee, J. M., Park, T. S., Oh, P. J., Lee, S.H., Lee, M. K., 2011. Application of digital sampling techniques for $4\pi\beta(\text{LS})-\gamma$ coincidence counting. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 626-627, 72-76.
3. Lee, K. B., Lee, J. M., Park, T. S., Hwang, H. Y., 2004. Implementation of tdc method in kriss. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 534 (3), 496-502.
4. Lee, K. B., Lee, J. M., Park, T. S., Lee, S.H., 2010a. Construction of classical confidence regions of model parameters in nonlinear regression analyses. Applied Radiation and Isotopes 68 (7-8), 1261-1265, proceedings of the 17th International Conference on Radionuclide Metrology and its Applications (ICRM 2009).
5. Michotte, C., Park, T. S., Lee, K. B., Lee, J. M., Lee, S.H., 2012. Comparison of ^{99m}Tc activity measurements at the kriss using the new sirti of the bipm. Applied Radiation and Isotopes 70 (9), 1820-1824.