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Primary standardization and Monte Carlo modelling of (^{243}Am + ^{239}Np) by two methods: TDCR and 4π(LS)-gamma coincidence counting system

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Abstract

The procedure followed by the Nuclear Metrology Laboratory (LMN) at the Nuclear and Energy Research Institute (IPENCNEN/SP), in São Paulo, Brazil, for the primary standardization of a (^{243}Am + ^{239}Pu) solution by two different methods, namely: TDCR and 4π(LS)-beta-gamma, is described. This work is complementary to the one presented at the ICRM'23 [*], where a proportional counter in 4π geometry was applied and using the same master solution.

The radionuclide ^{243}Am decays almost 100% by alpha transitions (with a very small branch, 10^{-9} %, of spontaneous fission) to ^{239}Np, in secular equilibrium, with a Q-alpha value of 5438.8 keV, and a half-life of 7367 years. Most of the decay (86.74 %) populates the excited level of ^{239}Np, with energy of 74.66 keV. The alpha decay is followed by a beta decay from ^{239}Np to ^{239}Pu, with a Q-beta of 722.5 keV, and a half-life of 2.356 days, populating mainly the excited levels of 228.18 keV (38.6 %) and 277.59 keV (34.8 %) of ^{239}Pu.

The measurements were carried out by a hybrid system composed of a TDCR and a 4π(LS)-beta-gamma coincidence counting system, where the gamma-rays are detected by a HPGe detector, placed below the TDCR system. The standardization was accomplished by dropping known aliquots of the solution into an Ultima Gold vial to be measured in the system. This master solution was the same as the one used in the previous calibration, performed in a 4π(PC)-beta-gamma coincidence system, providing direct comparison to the previous calibration.

The measurement was performed by a Software Coincidence Counting system composed of CAEN fast electronic modules, and an acquisition software developed at the LMN. These electronics provided individual amplitude and time stamps from the four acquisition lines: 3 PMT's and the HPGe.

This system provided the PMT's information for the TDCR analysis, as well as the N-(alpha+beta), N-gamma, and Nc counts, to be used in the coincidence equations, for two gamma gates: one at 75 keV, corresponding to alpha decay and another one at (228+278) keV, corresponding to beta decay. A dedicated software was developed for the analysis of the experimental data. Cox-Isham formalism was applied to experimental data and all partial uncertainties were considered, applying the covariance analysis methodology.

The experimental results were compared with Monte Carlo calculations, applying the code MCNP6, that considered all aspects of the counting system. A new analysis code was developed and applied, in order to calculate the alpha, beta, gamma and coincidence spectra, as well as the extrapolation curves, taking into account the decay scheme information. This provided an independent comparison with the experimental results.

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