

The Establishment of $4\pi\gamma$ Activity Measurement System Based on Well-type NaI(Tl) Detector Utilizing Monte Carlo Method

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Abstract

Background The well-type NaI(Tl) detector, a conventional $4\pi\gamma$ counting system, facilitates rapid sample screening but is constrained by efficiency calibration dependencies on standard sources and suboptimal geometric configurations. These limitations hinder measurements of irregularly shaped or difficult-to-prepare radioactive sources and necessitate efficiency corrections for cascade γ -emitting nuclides. **Objective** This work addresses these challenges by integrating Monte Carlo (MC)-simulated efficiency calibration with an optimized detector design. **Method & Results** Geant4 simulations determined the optimal well depth (85 mm for $\Phi 20$ mm diameter within a $\Phi 150 \times 150$ mm crystal) by analyzing efficiency curves of 14 nuclides and 31 mono-energetic γ -rays. Post-fabrication tests using a ^{137}Cs standard source validated critical performance metrics: background count rate ($n_b=109.7 \text{ s}^{-1}$), energy resolution ($\text{RE}=9.4\%$ @ 661.7 keV), and 12-hour stability ($\delta < 1.5\%$). Activity determinations for six nuclides (^{18}F , ^{22}Na , ^{68}Ge - ^{68}Ga , ^{133}Ba , ^{64}Cu , ^{198}Au) exhibited agreement ($|\text{En}| < 1$) with $4\pi\beta\text{-}\gamma$ coincidence and TDCR-LSC reference values. **Conclusion** The geometric optimization based on MC simulation made the detector close to the theoretical optimal configuration and reduced the uncertainty of efficiency correction while achieving relative expanded uncertainty $U_{\text{rel}} < 2.0\%$ ($k=2$), improving the work efficiency of radioactive source activity measurement. The reliability of the model was confirmed by measurement and comparison, and it can be used to simulate the detection efficiency of difficult to prepare or irregular radioactive sources, and provide reference for related research.

Full Text

Preamble

The Establishment of a 4 Activity Measurement System Based on a Well-type NaI(Tl) Detector Utilizing the Monte Carlo Method

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Abstract: [Background] The well-type NaI(Tl) detector, a conventional 4 counting system, facilitates rapid sample screening but is constrained by efficiency calibration dependencies on standard sources and suboptimal geometric configurations. These limitations hinder measurements of irregularly shaped or difficult-to-prepare radioactive sources and necessitate efficiency corrections for cascade γ -emitting nuclides. [Objective] This work addresses these challenges by integrating Monte Carlo (MC)-simulated efficiency calibration with an optimized detector design. [Method & Results] Geant4 simulations determined the optimal well depth (85 mm for a $\Phi 20$ mm diameter within a $\Phi 150 \times 150$ mm crystal) by analyzing efficiency curves of 14 nuclides and 31 mono-energetic γ -rays. Post-fabrication tests using a ^{137}Cs standard source validated critical performance metrics: background count rate ($n_b = 109.7 \text{ s}^{-1}$), energy resolution ($R_E = 9.4\% @ 661.7 \text{ keV}$), and 12-hour stability ($\delta < 1.5\%$). Activity determinations for six nuclides (^{18}F , ^{22}Na , ^{68}Ge - ^{68}Ga , ^{133}Ba , ^{64}Cu , ^{198}Au) exhibited agreement ($|E_n| < 1$) with 4 - coincidence and TDCR-LSC reference values. [Conclusion] The geometric optimization based on MC simulation made the detector close to the theoretical optimal configuration and reduced the uncertainty of efficiency correction while achieving relative expanded uncertainty $U_{rel} < 2.0\%$ ($k = 2$), improving the work efficiency of radioactive source activity measurement. The reliability of the model was confirmed by measurement and comparison, and it can be used to simulate the detection efficiency of difficult-to-prepare or irregular radioactive sources, providing reference for related research.

Keywords: radioactive activity measurement, NaI(Tl) scintillation detector, Monte Carlo method

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1. Introduction

The NaI(Tl) crystal scintillation detector is one of the most classic γ -ray activity measurement devices. This crystal features high density ($\rho_{\text{NaI}} = 3.607 \text{ g/cm}^3$)

and a relatively high average atomic number, resulting in strong attenuation power and high detection efficiency for γ -rays, along with good relative luminous efficiency, crystal transparency, and energy resolution. A cylindrical crystal can be machined into a well-type configuration by drilling a hole along its central axis, and the detector based on this crystal geometry is called a well-type NaI detector, which represents a common type of 4π counting detector. Its open structure allows virtually any type of γ -radiation source to be measured simply by preparing the sample as a small-volume source or placing it into a vial and inserting it into the well [1,2].

Due to the modest energy resolution of NaI detectors, activity measurements using well-type NaI detectors generally rely on the 4π full-energy spectrum counting method. This method extends the statistical range to the entire energy spectrum, using only a detection threshold to remove very-low-energy noise and interference, which makes it ideal for measuring nuclides with complex γ -ray cascades or high positron emission probabilities because of their high γ -ray emission rates. The simple principle of this method makes sample preparation and data processing very easy and fast. After efficiency calibration, the activity of radioactive samples can be obtained by combining the detection efficiency with information on emission probabilities and branching ratios from the decay scheme [3,4].

However, obtaining detection efficiency through traditional calculation methods is very complicated. In recent years, the Monte Carlo method (MCM) based on computer simulation has been widely used to perform efficiency calibration, with simulated values compared against reference values obtained from standard sources or devices to validate the accuracy of the simulation results [5].

To meet the need for rapid radionuclide measurement in the modern era, this work constructed a new well-type NaI detector and verified its measurement accuracy through comparison of simulated values with reference values. The radionuclides measured by this detector include ^{18}F , ^{22}Na , ^{64}Cu , ^{68}Ge , ^{133}Ba , and ^{198}Au , while the standard measurement devices that provided reference values include a 4π (PC)-(NaI) coincidence activity measurement device and a triple-to-double coincidence ratio (TDCR) liquid scintillation counter (LSC).

Following this verification, the 4π well-type NaI detector using detection efficiency obtained through the MCM can be deployed, improving the efficiency of activity measurements for common γ -radionuclides and positron-emitting nuclide samples, and laying the foundation for developing future standardized devices.

2.1 Efficiency Calibration and Monte Carlo Method

detectors require calibration to determine their detection efficiency before sample activity measurements can be performed. Calibration methods are classified into three categories based on the requirement for standard sources: active (source-based), passive (source-free), and hybrid (active-passive combined) [6].

Active efficiency calibration relies exclusively on standard sources, providing the highest accuracy and traceability reliability. However, this approach has two key limitations: dependence on physical detector-source compatibility and inapplicability to pre-construction stages. Specifically, it requires a fully constructed detector paired with specific standard sources, which necessitates a complex, time-consuming, and often costly source production process. Additionally, it cannot be implemented during theoretical studies or detector design phases before a physical prototype is available.

Passive efficiency calibration is typically performed via computer simulations using the MCM. The MCM is a computational approach widely used in radiation metrology and particle transport modeling. Its core principle is to estimate statistical expectations by simulating a large number of stochastic particle interactions [7]. MCM-based software tools (e.g., Geant4, MCNP, or EGS) simulate particle transport processes by constructing a geometric model of the detector and defining initial particle parameters (e.g., type, energy, direction), thereby generating simulated detection efficiencies [8].

The advantage of passive efficiency calibration is that it eliminates the need for physical detectors or standard sources, thereby reducing experimental preparation efforts. In simulated environments, both detector geometry and source parameters can be flexibly modified to meet new requirements, enabling high-statistics simulations through automated batch processing. This rapid and scalable approach addresses the growing demand for efficient calibration in modern radionuclide metrology [9,10].

The disadvantage of passive efficiency calibration is the absence of experimental validation, leading to uncertainties in model accuracy and result reliability. Therefore, passive calibration must be validated against active calibration once a physical detector is constructed. If simulated values (from MC simulation) agree with reference values (from experimental data on standard devices) across diverse scenarios (e.g., varying source geometries or energies), the model's predictive capability is deemed sufficiently reliable for subsequent applications [9,10].

In this work, Geant4 (version 11.2.1) was employed to construct the physical model of the well-type NaI detector [5,11]. By simulating the detection efficiency of various radionuclide sources (e.g., ^{22}Na , ^{60}Co , ^{137}Cs , ^{198}Au , and ^{241}Am) and mono-energetic γ -photon sources spanning (60–1860) keV, the optimized crystal geometry and detection efficiency for target radioactive sources were derived. The detailed detector parameters and simulation configurations are provided in Subsection 2.2.1.

2.2.1 Detector Geometry Optimization via MC Simulations

The scintillation crystal, as the core component of the NaI(Tl) detector, governs both its physical dimensions and detection efficiency. Compared to cylindrical geometries, the well-type configuration enhances detection efficiency by maximizing the solid angle subtended by the sample, albeit at the cost of reduced

spectral resolution due to light collection inhomogeneity. To leverage its intrinsic advantages, the well-type NaI detector developed in this work is optimized for maximum detection efficiency, the key prerequisite for implementing the 4 counting method [1-4].

The geometric parameters of a well crystal include the diameter/height of the crystal and the diameter/depth of the well. The crystal diameter/height upper limit is defined by manufacturing capabilities, and the well diameter lower bound is dictated by sample vial dimensions; these represent practical constraints. As for the well depth, it involves balancing two competing factors: solid angle and photon escape losses.

A deeper well can increase the solid angle (approaching 4π), improving efficiency, but excessive depth leads to γ -photon escape through the crystal base, which reduces efficiency.

To resolve this trade-off, MC simulation was employed to determine the optimal well depth for a given crystal diameter, height, and well diameter, thus establishing the geometric parameters of the detector model. This calculation process was referred to as detector geometry optimization (DGO) in this work. Its core is the calculation of crystal well depth based on MC simulation.

Based on the above theoretical requirements, data from existing cylindrical NaI detectors, and factory survey results, the material and geometry data of the well-type NaI detector (excluding well depth) were determined as shown in Table 1.

The model built in Geant4 based on these data is shown in Fig. 1 [Figure 1: see original paper] (the well depth is set at 85 mm tentatively).

To implement detector geometry optimization (DGO), 19 simulation models were established with well depths ranging from 30 mm to 120 mm in 5 mm increments. For each model, a source point was defined 1 mm above the well bottom along the central axis, containing 14 nuclide sources (listed in Table 2 with their γ -ray energies and emission probabilities [12]) and 31 single-energy γ -ray sources spanning 60 keV to 1860 keV in 60 keV increments. This energy range covers most characteristic γ -rays observed in practical applications. Each source emitted 10^7 initial particles, and simulated detection efficiency (ε_{sim}) was calculated as Eq. (1):

$$\varepsilon_{sim} = \frac{N_{threshold}}{10^7}$$

where $N_{threshold}$ represents counts above the 10 keV energy threshold in the simulated energy spectrum.

After the simulations were completed, efficiency curves for each nuclide and single-energy γ -ray source were generated across the tested well depths (30–120 mm). To resolve the trade-off between solid angle and photon escape, the

optimal well depth was determined by analyzing trends in all efficiency curves and selecting depths corresponding to peak values. Following this, the DGO was completed, and crystal processing could be initiated based on the optimized geometric parameters.

2.2.2 Detector Assembly and Testing

The NaI(Tl) crystal was machined into the well-type configuration specified by the DGO results and polished. Following this, the crystal, photomultiplier tube (PMT), and reflective materials were encapsulated in an aluminum alloy housing. A potential divider with safe high voltage (SHV) port and BNC signal output was subsequently installed externally to complete the detector assembly.

Two reflective materials were integrated into the design: MgO coated the crystal exterior, and PTFE lined the inner well walls, both enhancing light collection efficiency. Silicone gel served as the optical coupling agent between the crystal and PMT. The standardized dimensions of the NaI crystal ensured compatibility with commercially available PMTs. The ET9390KB model (ET Enterprises) was selected for its high pulsed linearity, pulse height resolution, and large light-receiving area ($\Phi 150$ mm), matching the crystal diameter.

Critical dimensions of the bare crystal and Al-alloy housing were verified against Table 1 specifications using calibrated, nationally traceable measurement tools. All measured deviations were confined to ± 0.02 mm, confirming manufacturing precision.

Fig. 2 [Figure 2: see original paper] depicts the assembly stages, from the drilled bare crystal to the encapsulated detector with a post-installed potential divider.

The encapsulated detector was stably housed in a 50 mm-thick cylindrical lead shielding chamber via a polyethylene support, which was positioned at the detector base, away from the crystal, to minimize interference with measurements. The assembled detector was first connected to a high-voltage power supply (Model PS350, SRS Inc.) and a multi-channel analyzer (MCA; DT5780P, CAEN) via the post-installed potential divider, then interfaced with a computer running MC2 Analyzer software, forming a complete 4 detection system. During subsequent testing and experiments, the detector operated at 650 V.

The performance of the assembled detector, including background count rate, energy resolution, and measurement stability, was evaluated using a ^{137}Cs point source under laboratory conditions, following standard testing protocols [13].

The γ -ray full-spectrum background counting rate (n_b, s^{-1}) is defined as:

$$n_b = \frac{N_b}{t_b}$$

where N_b is the total counts acquired in a background measurement over 10–30 minutes, and t_b is the live time (s). The design requirement is $n_b < 200 \text{ s}^{-1}$ in

formal measurements. To verify this, a baseline measurement of n_b must be conducted prior to housing the detector in the lead chamber, enabling quantitative assessment of its attenuation rate under the shield.

The detector energy resolution (R_E) is defined as:

$$R_E = \frac{FWHM}{D} \times 100\%$$

where R_E is the energy resolution for γ -rays of energy E (keV), $FWHM$ is the full width at half maximum (channel units) of the full-energy absorption peak (FEP), and D is the centroid channel number of the peak. To avoid the effects of statistical fluctuations, the peak count should be above 10^6 . For the well-type NaI detector, the resolution for the 661.7 keV γ -ray from ^{137}Cs was specifically validated, with a design requirement of $R_E < 10\%$. This value exceeds the typical 7–8% resolution of cylindrical NaI detectors due to the structural compromise inherent in the well-type design. As noted in Subsection 2.2.1, this trade-off was intentionally accepted to prioritize high detection efficiency.

The measurement stability of the detector is quantified by the relative drift (δ), defined as:

$$\delta = \frac{\Delta D}{D_0} \times 100\%$$

where ΔD is the peak position drift (in channel units) over the measurement period, and D_0 is the initial peak position. The δ represents the cumulative stability over this period. To evaluate δ , two measurements of the 661.7 keV γ -ray FEP were performed under continuous detector operation, separated by a prolonged interval. The design requirement is $\delta < 1.0\%$ for an 8-hour continuous run, or $\delta < 1.5\%$ for a 12-hour run.

The test protocol validates the detector's key performance parameters through three sequential measurements using a ^{137}Cs standard point source ($\sim 5 \times 10^3$ Bq). Initially, the well-type NaI detector was powered on and preheated at 650 V for 30 minutes. A 15-minute background measurement (N_b) was then conducted to calculate the background rate $n_b = N_b/t_b$ ($t_b = 900$ s), while simultaneously inspecting the energy spectrum for potential radioactive contamination. Following this, the ^{137}Cs source was positioned at the well bottom for the first standard measurement. Data acquisition continued until 10^6 counts accumulated in the 661.7 keV FEP (typically 3–4 minutes), allowing determination of the initial peak position (D_0) and FWHM via analysis software. To assess long-term stability, the detector remained operational at 650 V, with repeated standard source measurements at 8, 12, and 24-hour intervals. Peak drift ($\Delta D = D_t - D_0$) and relative drift (δ) were calculated for each interval, with design thresholds requiring $\delta < 1\%$ (< 6.6 keV drift) for 8-hour operation and $\delta < 1.5\%$ (< 9.9 keV) for 12-hour operation.

Upon completion of the test protocol, compliance with the specified thresholds for background count rate ($n_b < 100 \text{ s}^{-1}$), energy resolution ($R_E < 10\%$), and operational stability ($\delta < 1\%$ for 8 h) serves as validation of the detector's design and manufacturing quality. If all criteria are satisfied, the system is deemed suitable for laboratory-based radioactive source activity measurements.

2.2.3 Source Activity Measurement Based on Simulated Efficiency

The 4 counting method is implemented for activity measurements using the well-type NaI detector. This method is particularly suited for activity determination of cascade γ -emitting nuclides or positron-emitting nuclides with high γ -ray emission probabilities, provided the detector achieves near-4 π solid angles and detection efficiencies approaching 100% [3,4].

When the detector geometry and source characteristics satisfy the criteria of near-4 π solid angle and high detection efficiency, the full-spectrum net count rate ($n_0, \text{ s}^{-1}$) approximates the source γ -ray emission rate (N_0). The source-specific detection efficiency is defined as:

$$\varepsilon_s = \frac{n_0}{N_0}$$

representing the ratio of measured counts to emitted γ -rays. For a source with known nuclide decay scheme parameters (branching ratios and internal conversion coefficients), the activity ($A_0, \text{ Bq}$) is calculated via:

$$A_0 = \frac{n_0}{\varepsilon}$$

where $\varepsilon = \varepsilon_s(N_0/A_0)$ denotes the absolute detection efficiency, dependent on both detector response and the nuclide decay scheme. Given the sensitivity of ε to variables such as source geometry, γ -ray energy, and detector nonlinearity, calibration is mandatory.

As outlined in Section 2.1, MC simulations using Geant4 were employed to calibrate the detection efficiency (ε_{sim}) of the well-type NaI detector through passive (source-free) calibration, streamlining the efficiency calibration workflow. To validate this framework, active (source-based) calibration was subsequently performed by comparing simulated values against reference values obtained from certified standard devices [8-10]. The comparison focuses on the specific activity of the standard source solution used in source preparation.

Six kinds of calibration sources were chosen to cover a broad range of γ -ray energies and geometries: (1) Four positron emitters: ^{18}F (pure β^+ decay), ^{22}Na (β^+ decay with 1274.5 keV characteristic γ -rays), ^{64}Cu (three equilibrium decay pathways), ^{68}Ge - ^{68}Ga (parent-daughter system with complex emission

components); (2) ^{133}Ba (53–384 keV γ -rays) for low-energy validation; (3) ^{198}Au activated gold foil with uniform planar distribution to assess geometric response.

The ε_{sim} for all six kinds of sources should be obtained. For point sources, simulations of ^{18}F and ^{22}Na were completed during the DGO phase, while other nuclides were simulated under the optimized well depth model using identical protocols. ε_{sim} for these nuclides were derived from Eq. (1). The ^{198}Au activated gold foil simulation replaced the point source geometry with a planar configuration centered at the source point position, retaining all other parameters to ensure comparability.

Using ε_{sim} , the simulated activity A_{sim} was calculated as $A_{sim} = n_0/\varepsilon_{sim}$ (Eq. 7). The simulated specific activity S_{sim} (Bq/g) was then determined via $S_{sim} = A_{sim}/m$ (Eq. 8), with m (g) representing the mass of the source solution. While S_{sim} served as the primary comparison metric for point sources, the ^{198}Au activated gold foil retained A_{sim} for validation due to its special preparation method.

Having outlined the theoretical principles and experimental design of the 4 well-type NaI(Tl) detector, the following part details the standardized protocols for source preparation and measurement procedures. Identical protocols were applied to all five nuclides: ^{18}F , ^{22}Na , ^{64}Cu , ^{68}Ge - ^{68}Ga , and ^{133}Ba .

The original high-activity nuclide solution was first diluted to the target specific activity (100–500 Bq/mg), then sub-packaged into glass ampoules as reference standard solutions. Precise aliquots of the solution (~ 10 mg) were then dispensed via the differential-weighing method onto circular plastic support films ($\Phi = 18$ mm) to form 1–2 mm diameter source spots, with the mass of the solution used meticulously recorded. After air-drying, the sources were hermetically sealed, with three replicates prepared per nuclide (totaling 15 uniquely labeled sources) to ensure traceability. For short half-life nuclides such as ^{18}F ($T_{1/2} = 110$ min), strict time-sensitive protocols governing source preparation, measurement, and data acquisition are necessary to mitigate uncertainties from rapid decay.

The activated gold foil was fabricated by neutron irradiation of a pure gold foil ($\Phi = 15$ mm, thickness 100 μm) in a reactor, generating ^{198}Au uniformly distributed within the gold lattice [14]. This homogeneous distribution ensures the mechanical stability of the activated gold foil, resisting the detachment or contamination of ^{198}Au under contact or friction, thereby qualifying it as a pre-sealed planar source with dimensions comparable to sealed point sources. Measurement protocols for this planar source on the well-type NaI detector were identical to those for point sources, ensuring consistency across efficiency calibration workflows.

The prepared calibration sources were measured using the well-type NaI detector following a standardized protocol: background counts were first acquired, after which each source was positioned at the well bottom to initiate data acquisition once the count rate stabilized. Measurements were terminated when the full-

spectrum accumulated counts reached 10^6 , ensuring statistically robust results. Net count rates (n_0) were extracted via energy spectrum analysis by subtracting background contributions. Using the simulated detection efficiency (ε_{sim}) from Geant4, the activity of the source (A_{sim}) and specific activity of the solution (S_{sim}) could be calculated according to Eq. (7) & (8).

The final values of radionuclide solutions were actually determined by averaging the triplicate measurements of their corresponding sources, while the values of the activated gold foil were the average of its triplicate measurements. This process generated validated simulated values with their uncertainty that need to be evaluated for all six nuclides, enabling direct comparison with reference values to assess detector accuracy.

2.3 Measurement and Comparison of Reference Values

The simulated values derived from the well-type NaI detector (Subsection 2.2.3) require validation against certified reference values provided by primary activity standard devices. This section introduces the operational principles of two such devices—the 4 - coincidence activity measurement standard device and the triple-to-double coincidence ratio (TDCR) liquid scintillation counter (LSC) standard device—alongside their corresponding standard source preparation protocols. These devices serve as metrological benchmarks to independently verify the consistency between simulation-based results and experimentally traceable activity data.

2.3.1 4 - Coincidence Activity Standard Device

The 4 - coincidence activity standard device integrates a 4 proportional counter (PC, filled with argon-methane gas called P-10) and two cylindrical NaI γ -ray detectors positioned symmetrically. The 4 - coincidence method serves as an absolute measurement technique for β -cascade emitters (^{198}Au), X-cascade emitters (^{133}Ba), and positron-emitting nuclides (^{18}F and ^{22}Na) [15-17].

As the theoretical framework and methodology of the 4 - coincidence method have been rigorously validated in prior studies [18], this work directly applies its established principles and conclusions:

Under the experimental conditions here: (1) Both (PC) and (NaI) detectors exhibit near-uniform efficiency across mm-scale source spots. (2) Timing parameters (resolution time $\tau_R = 1$ s, dead time $t_D = 4$ s) and background shielding are properly configured. These conditions ensure reliable β -efficiency extrapolation, therefore the activity A_0 is determined via the relationship:

$$A_0 = \frac{n_{\beta 0} n_{\gamma 0}}{n_{co}} \cdot \frac{1 + \alpha}{1 + \alpha \varepsilon_{\beta\gamma} + \varepsilon_{ce}}$$

where $n_{\beta 0}$ is the β -channel net count rate, $n_{\gamma 0}$ is the γ -channel net count rate, n_{co} is the true coincidence rate, all in s^{-1} ; ε_{β} and ε_{γ} denote detection efficiencies of the corresponding channels; α is the internal conversion coefficient; $\varepsilon_{\beta\gamma}$ and ε_{ce} represent β -detector responses to γ -rays and conversion electrons.

To calculate A_0 , β -efficiency extrapolation is performed: varying ε_{β} generates data points for (x, y) where $x = (1 - \varepsilon_{\beta})/\varepsilon_{\beta}$ and $y = n_{\beta 0}n_{\gamma 0}/n_{co}$. Extrapolating the fitted curve to $x \rightarrow 0$ ($\varepsilon_{\beta} \rightarrow 1$) yields $y = A_0$. The A_0 was equal to the reference activity of the source (A_{ref} , Bq), from which the reference specific activity (S_{ref} , Bq/g) can be derived using the source solution mass.

After establishing the principles of the 4 - coincidence activity measurement method, the following part details the source preparation protocol and measurement procedures. The 4 - coincidence activity measurement protocol requires depositing standardized radioactive solutions (as detailed in Subsection 2.2.3) containing ^{18}F , ^{22}Na , and ^{133}Ba onto VYNS resin films coated with silicone-gold layers, creating 2–3 mm diameter source spots [19]. The differential weighing method dispensed approximately 10 mg of solution onto the film's center, which was subsequently air-dried. Ten replicates were prepared for each nuclide (50 sources in total) to ensure statistical validity and reduce defects caused by handling. The ^{198}Au activated gold foil was measured using the same protocol despite its planar geometry, as its sub-micron thickness had negligible impact on detection efficiency.

Before measurements commenced, the device was preheated and background radiation was recorded. Operational parameters were configured as follows: 4 -proportional counter voltage was set to 2650 V (using P-10 counting gas); β -NaI(Tl) detectors to 700 V; dead times to $t_D(\beta) = t_D(\gamma) = 4$ s; and the resolution time to $\tau_R = 1$ s. Each source underwent a 300-second measurement cycle, during which energy spectra were acquired simultaneously by all detectors.

Following data collection, the β -channel discrimination threshold was incrementally adjusted to produce multiple (x, y) datasets for efficiency extrapolation and to calculate the source activity. For the thin-film source nuclides (^{18}F , ^{22}Na , ^{133}Ba), A_{ref} was determined by averaging 10 replicate measurements. The A_{ref} for the ^{198}Au activated gold foil was obtained through 6 independent measurement repetitions. All four A_{ref} values and one S_{ref} value would be combined with their evaluated uncertainties to establish the final reference values.

2.3.2 Triple-to-Double Coincidence Ratio Liquid Scintillation Counter

The LSC is a widely used detector for low- to medium-energy β/γ particles, achieving near-100% photon detection efficiency through direct mixing of radioactive samples with liquid scintillator. This configuration provides enhanced sensitivity for low-energy radiation detection. The TDCR method is an absolute measurement technique implemented with LSC that determines sample activity through detection efficiency calculations derived from the ratio between triple and double coincidence events recorded by three PMTs [20]. ^{64}Cu exhibits

mixed decay modes with branching ratios of 17.52% (β^+), 38.48% (β^-), and 44.00% (electron capture, EC) [12]. All three decay pathways emit detectable electrons (particles or Auger electrons), which makes ^{64}Cu suitable for measurement using TDCR-LSC [21]. The ^{68}Ge - ^{68}Ga system faces challenges posed by its parent-daughter decay scheme, characterized by mixed β^+ /EC pathways and concurrent emission of low-energy X-rays during the decay process, making it also suitable for TDCR-LSC [22].

The TDCR-LSC consists of a cylindrical transparent vial containing a homogenized mixture of radioactive sample and organic scintillator cocktail, surrounded symmetrically by three PMTs arranged at 120° azimuthal intervals. Under idealized conditions with matched PMT detection efficiencies ($\varepsilon_1 = \varepsilon_2 = \varepsilon_3 = \varepsilon$), double-coincidence rate (n_D) and triple-coincidence rate (n_T) are measured through a coincidence circuit, interfaced with MCA and computational software [20,23].

The most simplified TDCR principle derives from binomial statistics: double-coincidence efficiency $\varepsilon_D = 2\varepsilon(1 - \varepsilon)$ and triple-coincidence efficiency $\varepsilon_T = \varepsilon^3$ establish the fundamental ratio $R = \varepsilon_T/\varepsilon_D = \varepsilon/(2(1 - \varepsilon))$. This monotonic relationship $R(\varepsilon)$ ($0 < \varepsilon < 1$) enables unique determination of detection efficiency through experimental measurements, allowing absolute activity calculation $A_{vial} = n_T/\varepsilon_T = n_D/\varepsilon_D$. In practice, due to differences in luminous efficiency among the three PMTs and their detection efficiencies for the continuous energy spectrum, ε_D and ε_T are expressed as free parameter functions $\varepsilon_D(\lambda)$ and $\varepsilon_T(\lambda)$, where λ represents the figure of merit (FOM) of the PMT system, defined as the particle energy required to generate a single photoelectron at the PMT photocathode. Consequently, the function $R(\varepsilon)$ transforms into $R(\lambda)$. The computational approach remains focused on solving the free parameter λ to calculate ε_D , ε_T , and ultimately A_{vial} . Once the vial source activity (A_{vial}) was determined, the specific activity of the radioactive solution (S_{ref}) could be calculated using the mass of solution dispensed during vial preparation [20,23].

In this study, TDCR-LSC was implemented to measure the specific activity reference value of ^{64}Cu and ^{68}Ge - ^{68}Ga solution. A quantified volume of scintillation cocktail (Ultima Gold-LLT, Perkin-Elmer Inc.; 12 mL) was added to an empty liquid scintillation vial, followed by precise dispensing of radioactive solution (~ 10 mg) via differential weighing. The mixture was homogenized by shaking to form a vial source (VS). Eight replicate VSs and one background control (non-radioactive) were prepared. Each VS was measured in the TDCR-LSC device (Hidex 300SL) to acquire N_T , N_D , and R values. Following application of the TDCR algorithm, the results from the eight measurements were averaged, with combined uncertainties evaluated to determine the final specific activity reference value (S_{ref}) for the nuclide solution.

2.3.3 Comparison of Measurement Results

After obtaining simulated and reference values for the specific activities of six nuclides, comparative analysis was performed to validate both the computational accuracy of the MC simulations and the reliability of the 4 well-type NaI detector.

The validation metric employed was the normalized error (E_n) calculated according to Eq. (10):

$$E_n = \frac{x_{sim} - x_{ref}}{\sqrt{U_{sim}^2 + U_{ref}^2}}$$

where x_{sim} denotes the simulated value, x_{ref} the reference value, U_{sim} the uncertainty of the simulated value, and U_{ref} the uncertainty of the reference value. The absolute normalized error $|E_n|$ determines compatibility between results: $|E_n| \leq 1$ means agreement within uncertainties, while $|E_n| > 1$ means statistically significant discrepancy [24].

For method validation, all six nuclides were required to satisfy $|E_n| \leq 1$. Successful compliance demonstrates that the well-type NaI detector system, when coupled with MC efficiency simulations, produces activity measurements consistent with standard reference methods within uncertainty bounds. This agreement confirms the technical feasibility and metrological reliability of the proposed 4 NaI detector/MC simulation approach for radionuclide standardization.

3.1.1 Well Depth Determined by Detector Geometry Optimization

As detailed in Subsection 2.2.1, 19 detector models with well depths ranging from 30 to 120 mm were constructed to optimize the detector geometry (DGO) of the NaI detector. Detection efficiencies were simulated for point sources of 14 nuclides (Table 2) and 31 mono-energetic γ -rays (60–1860 keV) across these depths to generate efficiency curves identifying the optimal well depth. Two data processing steps were implemented prior to analysis: (1) normalization accounting for varying γ -ray emission probabilities (4%–100%, Table 2), and (2) simplification of mono-energetic γ -ray efficiency curves.

For normalization, the detection efficiency at 75 mm well depth (the midpoint of both the 150 mm crystal height and the variable depth range of 30–120 mm) was set as unity for each nuclide, with efficiencies at other depths scaled proportionally. For mono-energetic γ -rays, an unweighted average of detection efficiencies across all 31 energies was calculated at each well depth. This produced a composite γ -ray detection efficiency curve as a function of well depth, which was subsequently normalized in the same manner.

Fig. 3 [Figure 3: see original paper] presents the processed efficiency curves. All mid/high-energy nuclides (including ^{137}Cs) and the averaged mono-energetic

curve exhibit peak efficiencies at 85 mm, with rapid declines at greater depths. Low-energy nuclides show inflection points at 85 mm, beyond which efficiency increases become negligible ($< 0.5\%$).

Consequently, the optimal well depth was determined as 85 mm ($\Phi 20 \times 85$ mm), achieving the best detection performance across the 14 nuclides and 60–1860 keV energy range. It should be noted that the conclusion “ $\Phi 20 \times 85$ mm well matches $\Phi 150 \times 150$ mm crystal” cannot be proportionally scaled. This arises from non-linear interactions between solid angle enhancement (initially proportional to depth but with diminishing returns) and photon escape reduction (governed by exponential attenuation). Larger crystals permit deeper wells due to reduced relative photon escape, yet even these exhibit an absolute depth limit as solid angle gains eventually plateau. Smaller crystals conversely restrict viable depths due to heightened escape probabilities.

3.1.2 Detector Performance Validation

Following installation (Subsection 2.2.2), the detector’s background count rate, energy resolution, and temporal stability were evaluated using a ^{137}Cs reference source under standardized protocols. Performance metrics derived from Eq. (2)–(4) demonstrated compliance with design specifications:

The shielded configuration achieved $n_b = 109.7 \text{ s}^{-1}$, representing a 90% reduction compared to unshielded conditions ($\sim 10^3 \text{ s}^{-1}$). This suppression limited background-induced uncertainty to 0.1% for sources at the $5 \times 10^3 \text{ Bq}$ activity level. Given the large-volume geometry of the NaI(Tl) scintillator, the measured n_b demonstrates exceptional performance, particularly considering the inherent challenges posed by its bulk dimensions in minimizing photon escape and environmental interference.

Energy resolution for the 661.7 keV γ -ray full-energy absorption peak measured $R_E = 9.4\%$, marginally exceeding conventional cylindrical NaI detectors yet remaining within the $< 10\%$ design requirement. Operational stability tests revealed peak centroid drifts $\delta = 0.9\%$ over 8 hours and $\delta = 1.3\%$ over 12 hours, satisfying the design requirements of $< 1.0\%$ (8 h) and $< 1.5\%$ (12 h). These results confirm the detector’s capability for prolonged high-precision measurements.

3.2 The Measurement and Comparison of Activity Values

According to Subsection 2.2.3, six radionuclides were involved in the (specific) activity measurement and comparison: (a) ^{18}F , (b) ^{22}Na , (c) ^{133}Ba , (d) ^{64}Cu , (e) ^{68}Ge - ^{68}Ga (initially as standardized solutions), and (f) ^{198}Au (initially as activated gold foil deposits).

In terms of simulated values (Subsection 2.2.3), the activity was determined via MC-simulated detection efficiencies in the well-type NaI(Tl) detector. For nuclides (a)–(e), simulated specific activities were calculated by averaging results

from three sealed point sources prepared from standardized solutions, with activities normalized to the solution mass. The simulated activity of (f) was derived from triplicate measurements of a single activated gold foil.

The reference specific activities of nuclides (a–c) were obtained by averaging measurements from ten thin-film sources (prepared from standardized solutions) using the 4 - coincidence system (Subsections 2.3.1). The TDCR-LSC measurements of eight liquid scintillation vials provided reference values for (d, e). Six repeated 4 - coincidence measurements on the activated foil yielded the reference activity of (f).

All values were decay-corrected to a common reference date and underwent full uncertainty evaluation. Key uncertainty components are detailed in Table 3 . Uncertainty components other than MC simulation efficiency were quantified following methodologies prescribed in standardized protocols [13,25]. The simulation efficiency uncertainty was derived from combined contributions of deviations across multiple approximate MC models and nuclear database inconsistencies.

Table 4 presents the final comparison results. The normalized error (E_n , Eq. 10) was calculated to assess consistency within uncertainty ranges. The results demonstrate $U_{rel} (k = 2) < 2\%$ for simulated activities of all six nuclides and $|E_n| < 1$ compared to reference values, confirming both the accuracy of the measurements and the consistency between simulated and reference values within their respective uncertainty ranges. This validates the 4 -NaI counting method using MC-simulated efficiencies against reference devices (4 - coincidence and TDCR-LSC) for these six nuclides with divergent decay characteristics, thereby verifying the method's feasibility and metrological accuracy.

4. Conclusion

This study successfully developed and validated a 4 activity measurement system based on a well-type NaI(Tl) detector through integrated MC simulations and experimental characterization. Key achievements include: (1) **Geometry Optimization:** Geant4 simulations of efficiency curves for multiple nuclides and mono-energetic γ -rays established an optimal well depth of 85 mm ($\Phi 20$ mm diameter) for a $\Phi 150 \times 150$ mm NaI(Tl) crystal, balancing photon absorption efficiency and geometric solid angle. (2) **Performance Validation:** Post-fabrication tests with a ^{137}Cs point source confirmed compliance with critical specifications, including background ($n_b = 109.7 \text{ s}^{-1}$), energy resolution ($R_E = 9.4\% @ 661.7 \text{ keV}$), and temporal stability ($\delta < 1.5\%$ over 12-hour operation). (3) **Metrological Accuracy:** Activity determinations (simulated values, with $U_{rel} < 2\%$, $k = 2$) for six nuclides (^{18}F , ^{22}Na , ^{68}Ge - ^{68}Ga , ^{133}Ba , ^{64}Cu , and ^{198}Au) demonstrated agreement ($|E_n| < 1$) with reference values from 4 - coincidence and TDCR-LSC activity standard devices, validating the MC-based efficiency calibration method across diverse decay schemes.

The geometric optimization of the detector based on MC simulation makes the

design close to the theoretical optimum and reduces the uncertainty caused by efficiency correction in subsequent measurement processes. The integration of MC efficiency modeling with a 4 well-type NaI detector enables rapid sample activity measurement and provides a feasible efficiency acquisition scheme for measuring irregularly shaped or difficult-to-prepare radioactive sources. This approach significantly enhances operational efficiency and scope of application in radionuclide metrology while maintaining traceability to national standards.

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Author Contributions: (a) Ling-Ze Meng: Conducted Monte Carlo simulations, prepared and measured sealed point sources (^{18}F , ^{22}Na , ^{133}Ba , ^{198}Au), performed data analysis and curation, and drafted the manuscript. (b) Bao-Ji Zhu: Prepared and measured radioactive sources for all six nuclides, managed standard reference devices, and contributed to data processing. (c) Ye-Hui Tian: Prepared and measured vial sources (^{64}Cu , ^{68}Ge - ^{68}Ga) and participated in data analysis. (d) Shun-He Yao: Designed experimental protocols and coordinated collaborations with manufacturers. (e) Wen Xia: Prepared and measured thin-film sources for 4 - coincidence measurements. (f) Li-Jun Xu: Oversaw experimental planning and project management.

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References

- [1] ZHANG Ming, YANG Yuandi, LIU Haoran. Study and Preliminary Results of the 4 Well-type NaI(Tl) Crystal Detector[J]. *Acta Metrologica Sinica*, 2014, 35(03): 296-299. (in Chinese)
- [2] LI Qi, CHANG Yongfu, XIE Feng, et al. Efficiency Calculation and Measurement of Large Volume Well-type NaI(Tl) detector[J]. *Atomic Energy Science and Technology*, 2003, (03): 230-232. (in Chinese)
- [3] LIAN Qi, CHANG Yongfu, LI Qi, et al. Specific Activity Measurement of ^{166m}Ho Solution by 4 Counting Method [J]. *Atomic Energy Science and Technology*, 2003, (04): 306-309. (in Chinese)
- [4] Ratel G. Activity Concentration of a Solution of ^{133}Ba : Results of an International Comparison [J]. *Nucl Instrum Methods*, 1992, A316: 318-323.
- [5] Allison J, Amako K, Apostolakis J, et al. Recent developments in Geant4[J]. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 2016, Vol.835: 186-225.
- [6] ZHU Wenkai, CHEN Jun, XIONG Jianping, et al. Test of accuracy for ISOCS measurement method by efficiency calibration without a radioactive source[J]. *Nuclear Electronics & Detection Technology*, 2005, (03): 287-290. (in Chinese)
- [7] XU Shuyan. The Application of Monte Carlo Methods in Experiment

- Nuclear Physics[M]. Beijing: Atomic Energy Press, 1996.
- [8] ZHANG Ming, WANG Renzhong, YANG Diyuan, et al. Influence of the Source Position to the Well-type NaI(Tl) Detection Efficiency Based on a Monte Carlo Simulation [J]. Nuclear Electronics & Detection Technology, 2012, 32(01): 50-53. (in Chinese)
- [9] GUAN Wei, BAI Wanchun, YANG Jing. Detection Efficiency Calibration of HPGe Spectrometers: Experimental Measurements and Monte Carlo Simulations [J]. Nuclear Electronics & Detection Technology, 2016, 36(02): 137-139+149. (in Chinese)
- [10] Saegusa J, Kawasaki K, Mihara A, et al. Determination of gamma-ray efficiency curves for volume samples by the combination of Monte Carlo simulations and point source calibration[J]. Journal of Nuclear Science and Technology, 2000, 37, (12): 1075-1081.
- [11] Agostinelli S, Allison J, Amako K, et al. Geant4—a simulation toolkit[J]. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2003, 506(3): 250-303.
- [12] LNE-LNHB/CEA. Mono BIPM-5 – Table of Radionuclides, Vol.8 [DB]. Bureau International Poids Mesures, Sèvres, France, 2016[2024-11-05]. <http://www.lnhb.fr/home/nuclear-data/nuclear-data-table/>
- [13] State Administration for Market Regulation. Calibration Specification of Ray Spectrometers of Scintillation Detectors: JJF 1744-2019[S]. Beijing: China Standard Press, 2019.
- [14] XU Xin. Theoretical Calculation of ^{198}Au and ^{199}Au Yields and Optimization of Irradiation Conditions[J]. Journal of Nuclear and Radiochemistry, 1980, (02): 100-106. (in Chinese)
- [15] GU Zhenjun, ZHANG Yiyun. 4 - Coincidence Counting for Absolute Activity Determination of ^{198}Au [J]. Nuclear Electronics & Detection Technology, 2008, 28(05): 1034-1036+1050. (in Chinese)
- [16] Ravindra Anuradha, Kulkarni D B, Sathian V, et al. Standardisation of ^{133}Ba by efficiency extrapolation method and calibration of ionisation chamber[J]. Applied Radiation and Isotopes, 2021, 174(prepublish): 109744.
- [17] Yamada T, Kawada Y, Sato Y, et al. Standardization of ^{18}F using the 4 (+) integral counting technique[J]. Applied Radiation and Isotopes, 2008, 66(6-7): 909-913.
- [18] WU Zhihua, ZHAO Guoqing, LU Fuquan, et al. Atomic Nuclear Physics Experimental Methods[M]. 3rd ed. Beijing: Atomic Energy Press, 1996: 218-225. (in Chinese)
- [19] Volkar M A, Wolke R L. The Preparation and Measurement of Very Thin VYNS Films[J]. Review of Scientific Instruments, 1969, 40(6): 849-851.
- [20] Broda R. A Review of the Triple-to-Double Coincidence Ratio (TDCR) Method for Standardizing Radionuclides[J]. Applied Radiation and Isotopes, 2003, 58: 585-594.
- [21] Zimmerman B E, Cessna J T, Fitzgerald R. Standardization of $^{68}\text{Ge}/^{68}\text{Ga}$ Using Three Liquid Scintillation Counting Based Methods[J]. Journal of research of the National Institute of Standards and Technology, 2008, 113(5): 265-80.

- [22] Bergeron D, Cessna J, Fitzgerald R, et al. Standardization of ^{64}Cu activity[J]. Applied Radiation and Isotopes, 2018, 139: 266-273.
- [23] LYU Xiaoxia, CHEN Xilin, YAO Shunhe. Development of TDCR Liquid Scintillation System [J]. Atomic Energy Science and Technology, 2014, 48(11): 2133-2136.
- [24] State Administration for Market Regulation. Rule for the Examination of Measurement Standards: JJF 1033-2023[S]. Beijing: China Standard Press, 2023.
- [25] General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China. Calibration Specification for Liquid-scintillation Counting System: JJF 1480-2014[S]. Beijing: China Quality Inspection Press, 2014.

Figures & Tables

Figure 1 is the visualization result of the Geant4 model of the detector. Its size and color were set according to the contents in Table 1. The well depth was 85 mm.

Fig. 1. The visual physical model of the detector (well depth is tentatively 85 mm)

Figure 2 is a collage of five photos of the detector manufacturing process, depicting the crystal cutting and measurement, crystal processing results, crystal packaging results, and two views of the finished detector in order.

Fig. 2. The photos of NaI(Tl) detector

Figure 3 shows the variation curve of simulated detection efficiency with well depth, including the detection efficiency of 14 nuclides and the average detection efficiency of 31 single-energy γ -rays in the energy range of 60–1860 keV, normalized according to the value at 75 mm. Where a) is the overall trend and b) is the local amplification of key area.

a) Overall trend of efficiency curve

b) Key area of efficiency curve

Fig. 3. The efficiency curves of point sources at different well depth

Table 1 lists the parameters of the Geant4 modeling process, including structure name, structure material, structure size, and visualization model color.

Table 1. The structural parameters and materials of the NaI(Tl) detector

Detector structure	Material	Size parameter	Color in model
Crystal scintillator	NaI(Tl)	$\Phi 150 \times 150$	Green
Reflector of outer crystal surface (Top/Side/Bottom)	MgO	4.5/2.0/2.0	Yellow

Detector structure	Material	Size parameter	Color in model
Shell of outer crystal surface (Top/Side/Bottom)	Aluminum Alloy (2A12)	2.0/3.0/5.0	Magenta
Window to PMT	Silicone-Gel	$\Phi 128 \times 2.0$	White
Reflector of inner well surface	Poly-tetra-fluoro-ethylene (PTFE/Teflon)	-	-
Shell of inner well surface	Al-Alloy (2A12)	-	-

Table 2 lists the main parameters of the 14 nuclides involved in the simulation, including nuclide names, primary γ -energy and emissivity.

Table 2. The radionuclides used for simulation and their γ -ray parameters

Nuclide	Energy of γ -rays (keV)	Probability (%)
^{241}Am	59.5	35.9
^{109}Cd	88.0	3.6
$^{22}\text{Na}^*$	511.0	180.8
^{137}Cs	661.7	85.1
^{60}Co	1173.2, 1332.5	99.9, 100.0
^{54}Mn	834.8	99.9
^{85}Sr	514.0	98.4
^{88}Y	898.0, 1836.1	91.9, 99.2
^{134}Cs	604.7, 795.8	97.6, 85.5
^{152}Eu	121.8, 344.3, 1408.0	28.4, 26.6, 20.8
^{208}Tl	2614.5	99.8
^{226}Ra	186.2	99.9

*Positron nuclides, which emit positrons that annihilate to produce a pair of 511 keV γ -photons.

Table 3 lists the possible resulting uncertainty components in various activity measurement schemes, including the source of the uncertainty components, measurement methods involved, the category of evaluation methods (A/B), and the approximate range of component values.

Table 3. The uncertainty evaluation of (specific) activity measurement results

Uncertainty Component	Involved Process	Evaluation Method	(%)
Solution weighing	Source preparation using nuclide solution	Type A	0.1–0.3
Statistical fluctuation	All counting processes	Type B	0.1–0.5
Measurement repeatability	Average calculation of multiple measurements	Type A	0.2–0.8
Background	All measurement processes	Type B	0.05–0.2
Dead time	All measurement processes	Type B	0.1–0.3
Simulated efficiency	Simulated values calculation	Type B	0.5–1.0
TDCR efficiency	TDCR-LSC measurement	Type B	0.5–1.0

Note: Figure translations are in progress. See original paper for figures.

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