Atom Indonesia

Journal homepage: http://atomindonesia.brin.go.id

Calibration Model Development and Performance Evaluation of Portable Multichannel Analyzer for In-Situ Nuclear Forensics

M. S. F. Husein[1,2], S. Sihana[3*], R. M. Subekti[2], K. Indriana[2], E. Noerpitasari[4], S. Susanto[2], L. N. Thanh[5]

[1]*Master of Physics Engineering, Universitas Gadjah Mada, Yogyakarta 55281, Indonesia*
[2]*Directorate of Nuclear Facility Management, National Research and Innovation Agency, Serpong 15314, Indonesia*
[3]*Department of Nuclear and Physics Engineering, Universitas Gadjah Mada, Yogyakarta 55281, Indonesia*
[4]*Research Center for Nuclear Materials and Radioactive Waste Technology, National Research and Innovation Agency, Serpong 15314, Indonesia*
[5]*Vietnam Agency for Radiation and Nuclear Safety, Hanoi, 112300, Vietnam*

ARTICLE INFO

Article history:
Received 20 December 2024
Received in revised form 27 April 2025
Accepted 4 May 2025

Keywords:

Calibration model
Performance evaluation
Portable multichannel analyzer
Nuclear forensics
Uranium quantity

ABSTRACT

The rapid response to nuclear security incidents necessitates precise, field-deployable instrumentation capable of quantifying nuclear material. This study evaluated the Portable Multichannel Analyzer (PMCA) equipped with a NaI(Tl) detector as a first-responding tool for nuclear forensics. Tertiary standards of natural and depleted uranium (0.5–30 g) were measured under varying acquisition times (60–1200 s) to establish calibration curves and determine performance metrics. Key performance indicators, including Relative Standard Deviation (RSD), uncertainty, and detection limits, were compared across linear and polynomial models. The PMCA demonstrated high precision with RSDs <8% for uranium masses >3 g. Polynomial calibration curves achieved superior accuracy ($R^2 > 0.99$), significantly reducing uncertainty compared to linear models. Benchmark comparisons with HPGe spectrometry and destructive potentiometric titration validated PMCA measurements, showing <5% deviation in uranium concentration. The findings established PMCA's viability as a rapid, reliable instrument for in-field nuclear forensic investigations, bridging the gap between laboratory-grade systems and field-deployable solutions.

INTRODUCTION

Nuclear security is a set of technical measures to prevent, detect, and respond to malicious acts involving nuclear and radioactive material or facilities [1]. Nuclear security for Material Out of Regulatory Control (MORC) obligates States to have a robust system in responding to nuclear security events with the ability to identify, categorize, and analyze nuclear or radioactive material out of regulatory control using nuclear forensics [2]. A good nuclear forensics system also serves as a deterrence, and the analysis result can be used to identify deficiencies in the Nuclear Material Accounting and Control System or vulnerabilities at border checkpoints and physical protection systems [3,4]. Typical response plan involves analyzing nuclear material identity directly at the Radiological Crime Scene with in-situ gamma spectrometry using low resolution detectors such as NaITl. This initial data will be used to formulate a series of nuclear forensics analyses and used for future decision-making [5,6].

Recent advancements in nuclear forensics emphasize the growing need for portable measurement instruments that provide accurate data for nuclear material or radioactive identification. The Portable Multichannel Analyzer (PMCA), equipped with low to medium-resolution detectors such as NaI(Tl) or CZT, presents a promising solution due to its portability, ease of use, and rapid data acquisition capabilities. Studies have demonstrated that portable detectors are not only

*Corresponding author.
E-mail address: sihana@ugm.ac.id
DOI: https://doi.org/10.55981/aij.2026.1584

used for identifying or categorizing radionuclide and nuclear material but also for quantitatively determining their activities [7,8]. Several measurement factors, such as geometry, sample form, acquisition times, and method of data analysis, often present a challenge to acquire accurate data using in-situ measurement [9].

High-Purity Germanium (HPGe) spectrometry is the gold standard for laboratory-based analyses due to its high resolution; it lacks the mobility required for field use because of its cooling system needs. In contrast, low-resolution detectors such as Na(ITl) equipped in PMCA offer advantages in portability, size, and weight, enabling in-situ measurements by first responders. Despite these benefits, resolution limitations in NaI(Tl) detectors can lead to inaccuracies in calculating net peak areas, which hinder precise quantitative analysis. Recent studies have proposed advanced calibration techniques and comparative analyses of detection methods to address these challenges [10,11]. This limitation on low-resolution detectors pushes the need for higher resolution instruments, such as HPGe or CZT detectors, for accurately calculating nuclear material concentration, mass, and isotope composition [12-14].

This research evaluates the ability of the PMCA with NaI(Tl) detectors to quantitatively analyze uranium in a nuclear material powder sample, including calculating mass and concentration. The novelty of this research lies in developing a method for calculating uranium concentration and mass with PMCA and in the comparison with high-resolution HPGE and validation of the concentration with potentiometric destructive analysis. There are three methods that will be used and compared to calculate uranium mass, which are comparison formulas, also calibration model with linear and polynomial equations. PMCA's performance will be assessed through measurements of natural and depleted uranium tertiary standards across a mass range of 0.5 g to 30 g and varying measurement times (60 to 1200 seconds). This study aims to demonstrate that low resolution NaI(Tl) detector can achieve precision comparable to HPGe instruments with a proper method to be used in field applications. While other similar research has compared NaI(Tl) with HPGe detectors, they have typically focused on soil samples or employed tabletop instruments [15]. Other research also compared uranium concentrations using methods like ICP-MS, but only using HPGe as the gamma spectrometry detector [16]. Comparison research for various gamma spectrometry detectors has been done, but none have included destructive analysis to benchmark the results [17-20]. This research not only evaluates the performance of PMCA against other non-destructive methods but also examines its reliability as a deployable in-field instrument for nuclear forensics. Additionally, the study benchmarks the uncertainties in uranium concentration calculations and compares PMCA's performance against International Target Values (ITVs) set by the IAEA [21].

This research contributes to advancing nuclear forensics capabilities by developing a method that can be used directly for field-deployable gamma spectrometry. The results of this research will improve the extent of in-field analysis done in direct response to nuclear security events and highlight PMCA's potential to enhance nuclear forensics analysis through accurate, portable measurement technologies, enabling effective identification and analysis in the in-situ response action.

METHODOLOGY

Tertiary standard materials

A series of tertiary uranium standards was used to create the calibration curve, correlating uranium mass with the net count rate of gamma radiation energy of 900 to 1001 keV. The natural and depleted uranium powder are acquired from Cameco in the form of U_3O_8. The materials were weighed using a Metler Toledo analytical mass balance with 0.1 mg resolution and analysed using a T90 Titrator for Potentiometry titration to precisely measure the elemental weight of uranium. The tertiary standards are placed inside a low-density polymer tube with a diameter of 4.5 cm. The measured elemental weights for each standard are presented in Table 1.

Table 1. Tertiary standard uranium weight.

Natural uranium (g)	Depleted uranium (g)
0.5216	0.5342
1.0169	1.0166
2.7817	2.7810
4.9813	4.9800
6.9439	6.9422
9.9466	9.9441
14.8847	14.8809
19.8776	19.8725
24.8144	24.8081
29.9502	29.9427

Unknown sample materials

The unknown sample is composed of three items. The first item is natural U_3O_8 produced by the conversion process in the pilot conversion plant

from Cogemma yellow cake. The gross weight of unknown sample one is 15.223 g. The second unknown sample is natural uranium produced from a different batch of the conversion process, combined with <50% of standard ThO_2 from Merck. The gross weight of the second unknown sample is 15.808 g. The third unknown sample is depleted uranium from an unknown origin in the form of U_3O_8 powder with a weight of 11.110 g. The three unknown samples are placed in the same container type as the tertiary standard and shown in Fig. 1.

Fig. 1. Unknown sample.

Instruments

The instruments utilized for this research are an analytical mass balance, a Portable Multichannel Analyzer (PMCA) equipped with a sodium iodide (NaI(Tl)) detector, a desktop Multichannel Analyzer (MCA) including a High-Purity Germanium (HPGe) detector, and a T90 Titrator for potentiometric destructive analysis. The Mettler Toledo analytical balance has a 210-gram capacity. The Mettler Toledo T90 Titrator is used to benchmark the measurement results from both non-destructive measurements and to accurately determine uranium contents.

The primary tool used in this study is the SAM 940 Portable Multichannel Analyzer, which was designed for field measurements and has a 2x2-inch NaI(Tl) detector and 256-channel spectrum analysis capacity. First responders can perform in-situ analysis in response to a nuclear security incident using this small device's real-time gamma spectroscopy capabilities. Although the SAM 940 has a lower resolution than the HPGe detector, it provides important information and enables quick assessments. The analyzer instantly gives dose rate information and spectrum readings for each energy peak that corresponds to the detected radionuclide using the installed Eagle OS. The Quantum NaID tool enables the determination of net peak values at certain gamma energies using Region-of-Interest (RoI) analysis by analyzing spectral data in the N24 format. Figure 2 shows the PMCA instruments.

(a) (b)

Fig. 2. Portable multichannel analyzer (a) and direct reading window (b).

The High-Purity Germanium (HPGe) detector from Canberra is used for accurate gamma spectrometry analysis. The device can handle about 4096 channels for high-resolution spectrum capture with an excellent energy resolution of 0.2 keV, which enables exact isotope characterization and guarantees exact gamma-ray peak identification. Genie 2000 software will be used to process the spectrum in order to calculate ROI and net count. This system will be used to compare the performance of PMCA and the developed method with high-accuracy MCA and a method generally used to calculate uranium mass. The Tabletop HPGe MCA system is shown in Fig. 3.

Fig. 3. Table top MCA with HPGe detector.

Measurement of tertiary standard

Natural and depleted uranium standards were measured with different acquisition times from 60 to 1200 seconds, with five repetitions for each mass and time combination, to assess the PMCA's performance. The experiments used gamma energy from 920 to 1030 keV produced by Pa-234m produced from U-238 decay, hence representing the most abundant isotope [18]. In contrast, the 185 keV gamma signal, which is unique to U-235, accounts for less than 1% of total uranium, making it less

reliable for bulk uranium measurement in these standards. The 1001 keV peak was also selected as it represents secular equilibrium with its parent in natural uranium samples and can be used to calculate the quantity of Uranium in the sample [22,23].

Tertiary standard measurements are checked for deviation using a graph that depicts the connection between the specific activity of the measured uranium in each mass and the variation of acquisition time. The specific activity is determined with Eq. (1), where each measurement net count rate is divided by the uranium mass in each standard. Specific activity can be defined as a number that remains constant across all samples, regardless of sample mass or acquisition variance [24,25]. The specific activity is regarded as an absolute value for each radionuclide, and it should not be altered or changed as long as the uranium comes from the same batch of material.

$$Standard\ U\ Specific\ Activity = \frac{net\ count\ rate\ (CpS)}{Uranium\ mass\ (g)} \quad (1)$$

Concentration calculation with comparison formula

The comparison formula involves calculating the concentration of uranium in the unknown samples by directly comparing the net gamma-ray peak area to that of a standard uranium sample of known mass. For this research, the 15-gram standard for both natural and depleted uranium was selected as the reference point due to its moderate mass, which minimizes the relative uncertainty in measurement while providing a clear signal for analysis. The net peak area will be selected from the best energy peak from the first evaluation. The comparison formula is used as shown in equation Eq. (2) [12,23], where the ratio of the net peak areas of the unknown sample and the standard is used to estimate the uranium concentration of the unknown sample:

$$Sampe\ Mass = \frac{Net\ count\ asample}{Net\ count\ standard} . Standard\ mass \quad (2)$$

This method provides a simple yet effective way to estimate concentration, assuming consistent detector efficiency and minimal variation in background radiation between measurements.

Concentration calculation with calibration model

The net count rate of the 1001 keV gamma energy peak region was identified using the Quantum NaID program, which analyzed spectral data in N42 format and calculated the net count with a Region of interest around 920 to 1030 keV. The net count rate is used to create a graph between net count and uranium mass to create a calibration curve. Two models were created for the calibration curve: a linear calibration curve and a second-order polynomial calibration curve. The polynomial model was developed to incorporate any non-linear effect. The research model of cross-variating mass and acquisition time produces an extensive dataset of 250 data points for each natural and depleted uranium category, ensuring abundant data for creating the calibration model.

The detection limits (LoD) and quantitation limits (LoQ) of the linear and polynomial models were calculated using the standard deviation of the lowest uranium standard and the slope of the calibration curve, which provides information on the smallest detectable and quantifiable uranium mass for each model [14,23]. Residual analysis was done after recalculating mass with both calibration models to compare the real mass of uranium standards against the residuals and determine if the model can be accurately used to calculate uranium mass [26,27].

The calibration curve was modeled using Python using the NumPy and SciPy libraries for linear and polynomial regression analysis. The linear model assumes a proportional relationship between mass and gamma signal, and the polynomial model accounts for potential non-linearities arising from self-absorption effects or slight variations in detector efficiency across the mass range. Eq. (3) shows the linear relationship formula between net count and mass. Using the linear relationship between net count and mass will be the basis to calculate Limit of Quantitation and Limit of Detection with Eqs. (4) and (5).

$$Net\ Count = a\ .\ Mass + b \quad (3)$$

$$Limit\ of\ Detection = 3.3\ . \frac{\sigma\ of\ lowest\ mass}{slope\ linear\ graph} \quad (4)$$

$$Limit\ of\ Quantitation = 10\ . \frac{\sigma\ of\ lowest\ mass}{slope\ linear\ graph} \quad (5)$$

The measurement's standard deviation of the lowest mass on the uranium standard series is denoted by σ. The coefficient of determination (R^2) and residual analysis were used to assess the goodness-of-fit of both models to ensure the calibration properly reflected the data over the mass range. According to Eq. (3), the mass of uranium is the x-axis while the net count is the y-axis in the linear equation, which will also be used in the

polynomial equation. This axis was selected because the net count is the impacted variable and uranium mass is the dependent variable. The two models are differentiated by the slope value, where the linear model only has one slope value, but the polynomial model has two slope values (a_1 and a_2) [28]. The polynomial equation is shown in Eq. (6) and derived to Eq. (9) to calculate the mass through Eqs. (7,8).

$$netcount = a_1 x\ mass^2 + a_2\ x\ mass + b \quad (6)$$

$$a_1 x\ mass^2 + a_2\ x\ mass + (b - netcount) = 0 \quad (7)$$

$$(b - \ net\ count) = b' \quad (8)$$

$$mass = \frac{-a_2 \pm \sqrt{a_2{}^2 - 4a_1 b'}}{2a_1} \quad (9)$$

The uranium content of the unidentified samples was determined using the calibration model equation. The accuracy and uncertainty of the concentrations derived using the calibration models and comparison formula were compared to evaluate the best method to calculate uranium mass and concentration. Statistical uncertainty in the measurement of net count and propagating errors from the calibration model were combined to compute relative uncertainties. Residuals of uranium tertiary standard mass were computed using Eq. (10) [27] to evaluate model fit, especially at lower and higher mass ranges. The residual value was plotted against the real uranium standard mass (x-axis) throughout the mass range of the tertiary standard. This was done to see the most accurate model for both uranium categories.

$$Residual = mass_{calc} - mass_{real} \quad (10)$$

Measurement of unknown sample and analysis comparison

Three instruments were used for analyzing unknown uranium samples to guarantee thorough examination and benchmarking the results. The PMCA was used for the initial measurement, and the optimal acquisition duration was selected through the examination of tertiary standards. Both the Eagle OS program for direct reading and the Quantum NaID software for Region of Interest (ROI) analysis were used to process the spectral data. The activity of U-238, which predominates in uranium samples, was represented by the 1001 keV gamma energy peak, which was used to compute the net peak area. Three different formulas, the comparison formula, the linear equation, and the polynomial equation, were used to calculate the uranium content in the unidentified samples. The calculation results are compared with the measurement and calculation of uranium mass and concentrations by established methods using HPGe high-resolution gamma spectrometry and potentiometric titration with a T90 titrator. The unknown sample and one standard will be measured by HPGe in 3600 seconds, and the mass of uranium will be calculated using the comparison formula.

Uncertainty analysis of pmca measurement

This research assesses measurement uncertainties in accordance with IAEA guidelines from International Target Value (ITV) for Safeguarding Nuclear Material [21]. Several key categories for Non-Destructive Assay (NDA) are described by ITV, including mass fraction (uranium, thorium, or plutonium concentration), U-235 enrichment (uranium isotopic composition), and mass determination of U-235 or Pu using gamma rays. The category "mass of U-235" was chosen as the reference for this study because it is the most identical parameter measured by this research. According to ITV, the procedure for determining U-235 mass is done by identifying the %U-235 enrichment in the uranium and measuring the total uranium mass in the sample. The U-235 mass is then calculated by multiplying the total uranium mass by its %U-235 enrichment [21].

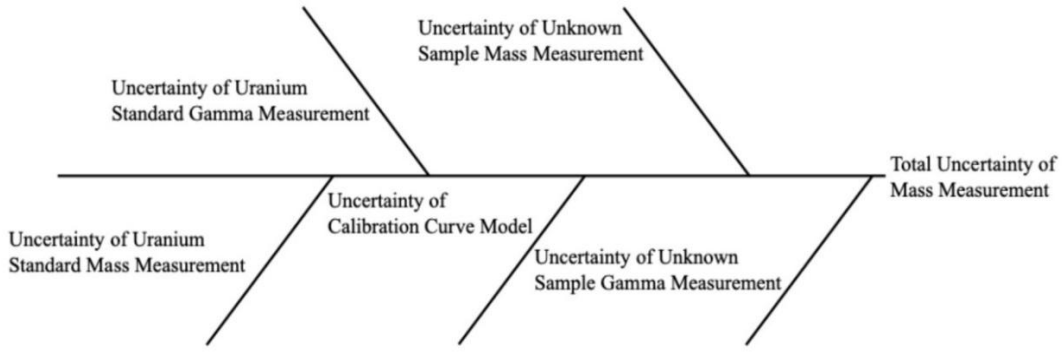


Fig. 4. Fishbone analysis for total uncertainty calculation.

The total uncertainty used to compare with the ITV is calculated following the fishbone diagram in Fig. 4 to incorporate various analysis steps. The standard deviation of repeated measurements for every sample and acquisition time was used to calculate random uncertainties. Systematic uncertainties were also incorporated, including those resulting from observational limits, net peak area extraction, and calibration model fitting. The Residual Standard Error (RSE) of the model shown in Eq. (11) is used to determine the calibration model's uncertainty [27].

$$RSE = \sqrt{\frac{\sum(y_{calc} - y_{real})^2}{N - p}} \qquad (11)$$

The top part of the equation is the value from residual in Eq. (10), N is the number of points in the calibration curve, and p is the number of parameters of the model formula. The linear model has a p of 2 because it only calculates slope and intercept, while the polynomial model has a p value of 3. N is 10 for the number of mass variations in the standard.

RESULTS AND DISCUSSION

The first data analysis was done to investigate the Relative Standard Deviation (RSD) of uranium measurements across varying acquisition times and mass variations to ensure the precision of measurement for both natural uranium and depleted uranium. Figure 5 shows the distribution of the RSD (Y) across acquisition time or live time variation (X) on different uranium masses, shown by different line styles.

The specific activity for each variation in the natural and uranium samples is presented in Fig. 6. The result of the measurement on each acquisition time variation is averaged to make the dataset simpler. Figure 7 graph shows the relationship between 1001 keV net count with uranium mass to describe the best model for quantifying uranium mass. The figure shows two calibration models with different lines. The graph in Fig. 7 serves to investigate how different trendlines correlate with the data produced from measuring the tertiary standard series and to extract the characteristic value from each calibration model.

Table 2 shows the characteristic values to complete the equation for calculating uranium mass. The slope (a) and intercept (b) values from Table 2 are used to complete Eqs. (3), (6), and (9).

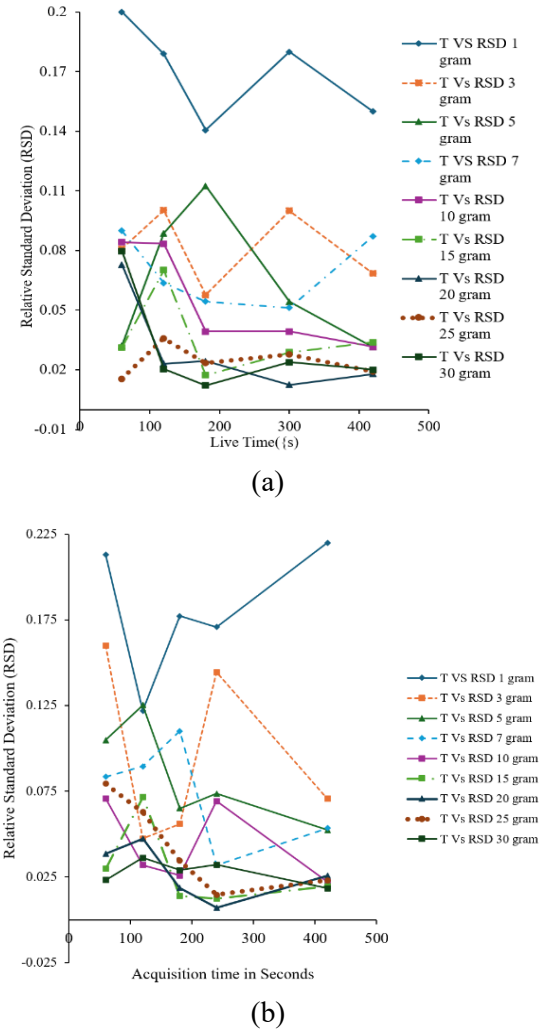
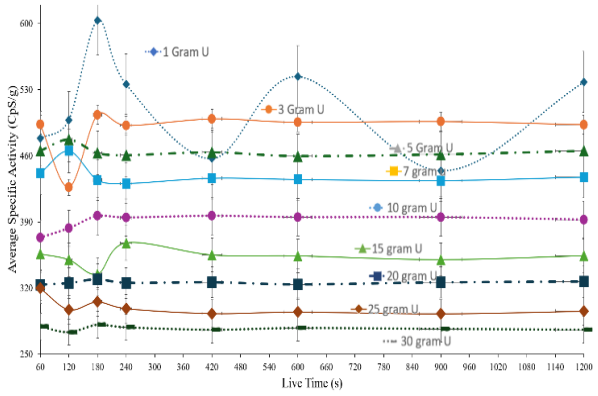


Fig. 5. Relative standard deviation distribution for time and mass variation of natural uranium (a) and depleted uranium (b).

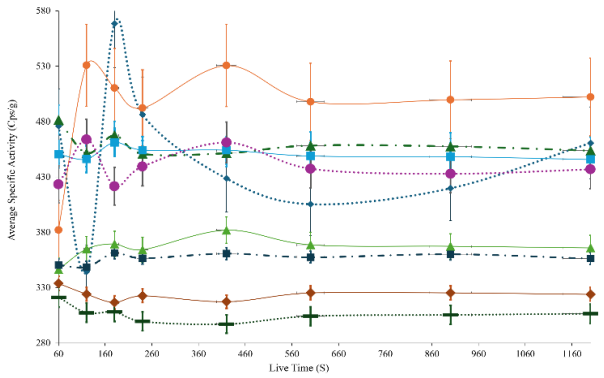
The equation is used to calculate the weight of uranium from each tertiary standard series according to linear and polynomial equations, and the result is used for residual analysis. The residual values are calculated using Eq. (10), and Fig. 8 shows the residual analysis result for both depleted and natural uranium.

Table 2. Measurement characteristic value from standard uranium.

Characteristic value	Natural uranium	Depleted uranium
Limit of Detection (g) linear	0.42	0.32
Limit of Detection (g) polynomial	0.27	0.21
Limit of Quantitation (g) linear	1.30	0.97
Limit of Quantitation (g) polynomial	0.81	0.62
a linear	276.58	295.76
b linear	686.21	811.83
R^2 linear	0.98	0.98
a_1 polynomial	-5.3344	-5.3125
a_2 polynomial	430.54	458.31
b polynomial	136.46	76.389
R^2 polynomial	0.99	0.99

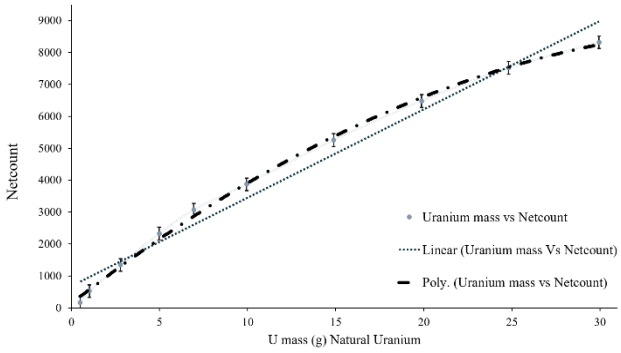


(a)

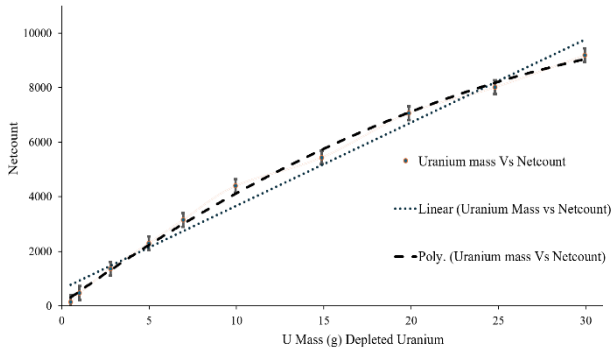


(b)

Fig. 6. Specific activity of each measurement variation for natural uranium (a) and depleted uranium (b).

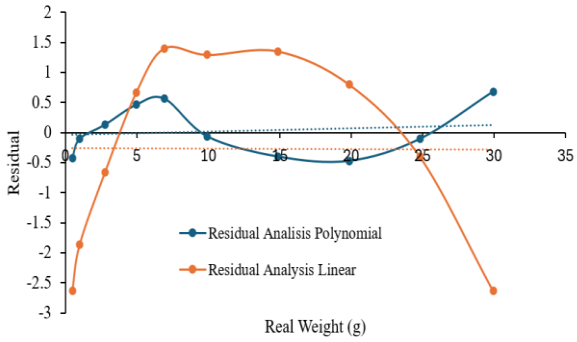


(a)

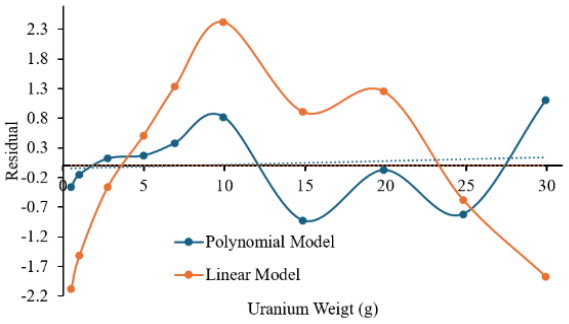


(b)

Fig. 7. Graph and trendline model of uranium mass vs net count for Natural Uranium (a) and Depleted Uranium (b).



(a)



(b)

Fig. 8. Residual analysis for linear and polynomial model for Natural Uranium (a) and Depleted Uranium (b).

PMCA performance

The first step of this research is to evaluate the precision of PMCA. Figure 5 shows the RSD value for different masses and acquisition times or detector live time. The figure shows that RSD is lower as the mass goes higher according to different line styles and colors, while all the lines do not have any specific pattern that proves time has no significant effect on RSD. This result is different from other research where acquisition time can affect the precision of gamma spectrometry measurement [8,10,18,19,29]. Uranium mass above 3 grams produces RSD values below 10%, while lower masses, such as 1 gram, exhibited higher values between 12% and 22% for both categories of uranium. Measurements for the 0.5-gram standard were excluded due to RSD exceeding 40%, to show a clearer graph in Fig. 5.

Specific activity results in Fig. 6 support the finding in Fig. 5. Both uranium samples with masses over 3 grams showed minimal deviation, with consistent activity values across acquisition times from 180 to 1200 seconds. The different values of specific activity on each line style demonstrate the effect of self-attenuation, where the mass of uranium attenuates the radiation from reaching the detector. Previous studies have quantified self-attenuation effects, incorporating correction factors into detector efficiency calculations [24,25,29], but this research

incorporates this phenomenon by using a polynomial calibration model to calculate uranium mass.

The second step of this research is to determine the optimal calibration model to calculate uranium mass from around 920 to 1030 keV net count. Table 2 shows that the linear model has a lower R^2 score than the polynomial model. This low R^2 value indicates the inability of the linear model to capture the non-linear relationship caused by self-attenuation effects, which become significant as uranium mass increases. This effect, caused by the increment of shielding from the material itself, reduces the total detectable radiation than expected. The polynomial model, with an R^2 value of 0.99, provides a better fit by incorporating this phenomenon. This model should accurately represent the relationship between uranium mass and net count, which will make the calculation result more accurate. The Limit of Detection (LoD) and Limit of Quantitation (LoQ) are calculated from the standard deviation of the 0.5-gram uranium tertiary standard and the slope of both calibration models. The polynomial model has lower LoD and LoQ, which means that this model not only can confirm the content of uranium in lower uranium mass, but it can also calculate accurately a lower mass of uranium than the linear model. Table 2 also shows the model characteristic values, which are the intercept and slope for each model on both uranium categories. The slope (a) value on each model for different uranium categories is quite similar, which means that natural uranium and depleted uranium have little effect on the 1001 keV gamma relationship with the mass of uranium. The intercept value, which some suggest affects the LoQ, is lower for the polynomial model, which supports the argument that the polynomial model can quantify uranium in lower mass than the linear model.

Figure 8 on residual analysis supports the argument that the polynomial model can more accurately calculate uranium mass than the linear model. Graph a on natural uranium shows that the linear model has a reverse parabolic pattern where the linear model underestimates uranium mass by 1 gram, then overestimates the uranium mass in the middle area of the model, and underestimates higher mass. This pattern indicates that the linear model will give a uranium mass value far from the actual value, where the polynomial model has no specific pattern, and the line is close to the middle line, which indicates there are little to no differences between the calculated mass and the real mass of uranium. Graph b on depleted uranium shows even more promising results with an oscillating line of the polynomial model that indicates the polynomial model for depleted uranium is more likely to achieve higher accuracy than the polynomial model on natural uranium. This makes the polynomial calibration curve a more suitable formula to calculate uranium mass. These results suggest a more accurate method than using the comparison formula, which is mainly used to calculate uranium mass in another research [23,30].

Unknown sample result and measurement benchmarking

The chosen acquisition time for this phase is 240 s because the RSD result is not much different from a longer acquisition time and because PMCA is intended to be used for in-situ analysis, which needs a faster time. The unknown samples are measured, and the spectrum is read with both Eagle OS and Quantum NaID, which show the same value for Uranium net count. The net count data were then used to calculate uranium mass and concentration with Eqs. (2), (3), and (9). Benchmarking was also done with HPGe and potentiometric measurement. Table 3 shows the results from unknown samples and uses potentiometric titration as the baseline for calculating the % error in both non-destructive methods.

The first phase of this research shows that PMCA has adequate precision in measuring natural and depleted uranium. This phase will discuss how its performance on analyzing and quantifying uranium in unknown samples using both calibration models that have already been discussed before, along with the comparison formula that is mainly used in another research.

Table 3 shows the overall result of this phase, where the three unknown sample uranium masses are calculated with three different formulas for PMCA measurement and benchmarked with results from HPGe and destructive analysis that are set as the true value. The PMCA and HPGe deviations for samples 1 and 3 were less than 1000 counts per second, indicating agreement between two different detectors. Sample 2 (U+Th combination) net count for PMCA and HPGe have big differences, with 19000 cps for PMCA and 3000 CPS for HPGe in the same energy peak. This implies that the radiation from thorium disrupts the 1001 keV energy gamma of Pa-234m, and due to the low resolution of PMCA, it cannot differentiate the radiation and therefore increases the net count on that energy.

Table 3 clearly shows that the argument of the polynomial model being the most accurate is true by showing the mass of uranium in samples 1 and 3 being close to the result acquired from potentiometry analysis. The difference between potentiometry analysis and PMCA measurement with a polynomial calibration model is less than 1 gram for natural uranium and less than 0.5 gram for depleted uranium.

Table 3. Unknown samples measurement result from PMCA, HPGe, and Potentiometry.

	Unknown Sample 1 (Natural U)	Unknown Sample 2 (Nat. U + Th)	Unknown Sample 3 (Depleted U)
Nett Weight Uranium + Impurity (g)	15.22	15.81	11.11
net count (PMCA)	4800.4	19170,5	3589.56
net count (HPGe)	4564	3014	3353
Mass: Comparison Formula PMCA(g)	13.60	54.33	9.82
Total Uncertainty comparison formula (%)*	1.14	-	2.96
Mass: linear equation PMCA (g)	14.60	66.51	9.73
Total Uncertainty linear model (%)*	25.72	-	29.77
Mass polynomial equation PMCA (g)	12.89	-	8.27
Total Uncertainty polynomial model (%)*	4.48	-	9.00
Mass from HPGe (g)	10.78	7.11	7.91
Total Uncertainty HPGe (%)*	0,05	0,07	0,03
Mass from Potentiometry (g)	11.91	8.44	8.62
Concentration comparison PMCA	89.36%	349.3%	88.41%
Concentration linear PMCA	93.96%	427.6%	87.55%
Concentration polynomial PMCA	84.69%	-	77.57%
Concentration HPGe	70.82%	45.02%	71.97%
Concentration Potentiometry	78.25%	54.24%	77.57%
Error in Concentration (PMCA)**	8.230%	>500%	<0.001
Error in Concentration (HPGe)**	9.49%	16.99%	7.21%

* % to the measured value, will be compared to the ITV total uncertainty of 6.7%

** Measurement error calculated from the closest value to potentiometry, that set as the true value

The linear calibration model and comparison formula show more than 2 g of difference on natural uranium and around 1 gram difference on depleted uranium. The HPGe measurement, on the other hand, shows a slightly higher error than PMCA for both samples. The HPGe measurement with a comparison formula to calculate uranium mass resulted in around 1.1 g difference on sample 1 and 0.8 g difference on sample 3, while sample 2, that previously cannot previously be determined with PMCA, is countable with HPGe with the mass difference from the potentiometry analysis of 1.3 g. This shows that HPGe has superior detectability than PMCA due to the detector's high resolution, but PMCA with a polynomial calibration model has higher accuracy. This argument is also supported by the data on errors in concentration for PMCA, where the error in concentration is less than 9% for sample 1 and less than 0.01% for sample 2, while HPGe has a higher error in the concentration.

The final verdict on this benchmark is that PMCA has higher accuracy when a polynomial model is used to calculate uranium mass due to the model incorporating the self-attenuation effect that makes the relationship between uranium mass and net count non-linear.

The nonlinear effect happens because as the mass of uranium increases, it changes the geometry of the whole sample inside the sample container. After all, the sample is in the form of powder with the same uranium concentration, so increasing the uranium mass means increasing the number of powders inside the container, therefore increasing the thickness of the sample inside the container. The HPGe detector has more accuracy and precision than PMCA if both detectors use the same method to calculate uranium mass, which is a comparison method; therefore, if the polynomial calibration model is also made for the HPGe, it will have higher accuracy than the PMCA.

Uncertainty comparison

The total uncertainty value in Table 3 is calculated differently for each method used to calculate uranium mass. The comparison formula is the simplest method where uncertainty only comes from measurement of one standard and the unknown sample and the calculation with calibration curve uses all step from measuring series of tertiary standard, calculating each standard mass and net count, to creating different calibration model therefore incorporating every element in the fishbone graph shown in Fig. 4. Table 3 shows that comparison formula have the lowest uncertainty in for PMCA measurement due to its simplicity while the polynomial model show the second lower uncertainty due to low Residual Standard Error (RSE) of the whole model. Linear calibration models have the highest uncertainty because they have a higher RSE value, which can be compared with the polynomial model from the residual analysis graph. The higher resolution of HPGe with 10 times longer acquisition time helps HPGe to achieve lower deviation in the measurement result [31,32]. HPGe also utilizes a comparison formula with fewer steps for calculating uranium mass, resulting in fewer components to the total uncertainty. The PMCA with a polynomial model achieves an uncertainty value of 4.48% for natural uranium and 9% for depleted uranium. The ITV for uncertainty in calculating Uranium mass is 6.7% [21], so PMCA achieves proper uncertainty for natural uranium but not for depleted uranium. Although depleted uranium has a more accurate result in this research, its precision must be improved for the measurement's result reproducibility, either by increasing the number of repetitions in the unknown sample measurement or by decreasing the RSE value of the calibration model with the tertiary standard.

Performance of PMCA to support nuclear forensics first responder

According to the International Atomic Energy Agency Implementing Guide No.22-G about Radiological Crime Scene Management, one of the main purposes of interfering with the crime scene is the identification of radionuclides, whether in their material form or as contamination [33]. The goal for the identification is to evaluate the risk associated with the material, determine whether there are any laws broken in the presence of the materials, and formulate further investigative procedures on nuclear forensics. In-field forensic investigation is sometimes done if it is requested by the investigator or command center, but it's usually for traditional forensics.

This research has proven that standard model portable multichannel analyzer, such as SAM940, that are not initially equipped with any multigroup analysis software or uranium quantitation software can be used to calculate uranium mass and concentration using a simple procedure. Preparing a calibration model for a specific instrument with a polynomial equation to calculate uranium mass is the only thing needed for standard field-deployable instruments, such as PMCA in this research, to accurately quantify uranium. This performance and novel calibration model can help acquire much more information on uranium, which is a nuclear material that is generally used for nuclear fuel and has the highest risk of being diverted [1]. These research findings can be used to improve nuclear forensics capabilities in developing countries such as Indonesia, where high-resolution portable gamma specs equipped with uranium quantity calculating software are still not generally available for first responders or present in limited numbers. A simple PMCA with this developed calibration model can be a dependable option to identify uranium in nuclear security events with high accuracy proven by this paper. Further research in this field can be focused on developing a method for calculating the isotopic composition of uranium for further identification and developing a method for quantifying uranium mixed with thorium, which this research is still unable to calculate.

CONCLUSION

The Portable Multichannel Analyzer (PMCA) with a NaI(Tl) detector is shown in this research to be a feasible, field-deployable instrument for in situ nuclear forensics analysis in responding to nuclear security events. The PMCA's accuracy was confirmed by measuring both natural and depleted uranium tertiary standards. For uranium masses more than 3 grams, the relative standard deviations were less than 10% for all acquisition times. When compared to destructive analysis (potentiometric titration), the polynomial calibration model demonstrated the highest accuracy in determining uranium mass and concentration, with an error of less than 0.01% for depleted uranium and just 8.17% for natural uranium. The measurement with the polynomial model also achieves a 4.48% uncertainty for natural uranium but a 9% uncertainty for depleted uranium. The measurement result for natural uranium satisfies the international target value (ITV) by IAEA, with the value below 6.7% but the depleted uranium performance still needs improvement, either by increasing the number of repetitions for unknown samples measurement or decreasing the RSE value of the calibration model through increasing the number of points on the tertiary standard.

Results of this research prove PMCA's capability to be used as in-situ nuclear forensics analysis equipment, serving as a dependable option for responding to nuclear forensics and used in radiological crime scenes. Further research on this subject can be focused on developing a method to quantify isotopic composition and calculate uranium or other nuclear material in mixed U-Th samples.

ACKNOWLEDGMENT

I would like to express my sincere gratitude to Badan Riset dan Inovasi Nasional (BRIN) for awarding me the Degree by Research scholarship, which enabled me to pursue my master's degree in physics engineering. This research is part of my thesis and would not have been possible without their generous support.

I would like to extend my sincere appreciation to the Coordinated Research Project on Nuclear Forensics Team (J02020) for their invaluable technical and financial support throughout the course of this research.

AUTHOR CONTRIBUTION

M. S. F. Husein, as the first author, conceptualized the research and developed the methodology. S. Sihana, the corresponding author, guided the overall direction of the research. R. M. Subekti, as the research supervisor, evaluated the technical methods, data, and data analysis process. K. Indriana and Susanto contributed to refining the methodology and processed the data to generate graphs and other valuable insights. E. Noerpitasari and L. N. Thanh co-authored the

introduction and discussion on nuclear forensics, with E. Noerpitasari also contributing to the HPGe measurement and the subsequent data processing for HPGe results.

REFERENCES

1. International Atomic Energy Agency (IAEA), Implementing Guide: Sustaining a Nuclear Security Regime, Nuclear Security Series No. 30-G, IAEA, Vienna (2018).
2. S. Maity, A. Pant, S. Police *et al*., Role of Nuclear Forensics in Preparedness and Response in Radiation Emergencies, in: Handbook on Radiation Environment, Vol. 1, Springer, Singapore (2024).
3. J. M. Schwantes, O. Marsden, and K. L. Pellegrini, J. Radioanal. Nucl. Chem. **311** (2017) 1441.
4. G. R. Eppich, Bull. At. Sci. **76** (2020) 263.
5. S. Mishra, S. Anilkumar, and A. Vinod Kumar, Nuclear Forensics: Role of Radiation Metrology, in: Handbook of Metrology and Applications, Springer, Singapore (2023).
6. W. Wang, J. Xu, R. Xi *et al.*, J. Anal. At. Spectrom. **39** (2024) 178.
7. P. E. Biere, J. O. Aina, M. O. Olaoye *et al.*, Mater. Geoenviron. **68** (2021) 1.
8. S. Guguła, K. Kozak, J. Mazur *et al.*, J. Environ. Radioact. **188** (2018) 87.
9. K. Tsuchiya, K. Kurosawa, N. Akiba *et al.,* Eur. Phys. J. Plus **133** (2018) 266.
10. H. El-Gamal, H. Negm, and M. Hasabelnaby, J. Radiat. Res. Appl. Sci. **12** (2019) 360.
11. A. R. A. Abdullah, S. Permana, W. Srigutomo *et al.,* J. Eng. Technol. Sci. **57** (2025) 105.
12. S. A. Okasha, A. A. Faheim, M. H. E. Monged, *et al.*, Int. J. Environ. Anal. Chem. **103** (2023) 737.
13. F. Salvador, T. Marchais, B. Perot *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **1065** (2024) 169549.
14. T. Marchais, B. Pérot, C. Carasco *et al.*, IEEE Trans. Nucl. Sci. **69** (2022) 761.
15. S. Maeng, H. Y. Lee, S. J. Park *et al.*, J Radioanal. Nucl. Chem. **331** (2022) 2929.
16. E. Chajduk and P. Kalbarczyk, Nukleonika **66** (2021) 121.
17. A. Mishra and R. Khanal, Atom Indones. **1** (2023) 109
18. R. Pradana, E. D. Nugraha, W. Wahyudi *et al.*, Environ. Sci. Pollut. Res**. 30** (2023) 89280.
19. W. J. C. Koppert, M. M. A. Dietze, S. van der Velden *et al.*, Phys. Med. Biol. **64** (2019) 135012.
20. D. Bednár, A. Pažitný, M. Ličtjak *et al.*, Ann. Nucl. Energy **159** (2021) 108335.
21. International Atomic Energy Agency, International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials, IAEA, Vienna (2022).
22. I. Meleshenkovskii, N. Pauly, and P. E. Labeau, Appl. Radiat. Isot. **148** (2019) 277.
23. D. Ko, S. Cheon, J. Kim, S *et al.,* Nucl. Eng. Technol. **57** (2024) 103156.
24. A. Serban, A. Apostol, L. Ganea, *et al.*, Rom. Rep. Phys. **73** (2021) 201.
25. E. O. Akinduro, Mod. Appl. Sci. **16** (2022) 4.
26. H. Fu, J. Jia, H. Wang, *et al.*, Calibration Methods of Laser-Induced Breakdown Spectroscopy, in: Calibration and Validation of Analytical Methods - A Sampling of Current Approaches, InTech., USA (2018).
27. D. Chen, F. Hu, G. Nian *et al.*, Entropy **22** (2020) 193.
28. M. Zehtabvar, K. Taghandiki, N. Madani *et al.*, Spectrosc. J. **2** (2024) 123.
29. I. M. Nabil, K. M. El-Kourghly, Y. Mohamed *et al.*, Radiat. Detect. Technol. Methods **8** (2024) 1641.
30. S. A. Abdualhadi, B. Mauz, S. D. T. Joss, *et al*., Radiat. Meas. **114** (2018) 25.
31. T. Marchais, B. Pérot, C. Caraso *et al.*, IEEE Trans. Nucl. Sci. **67** (2020) 654.
32. P. G. Allinei, N. Pérot, B. Pérot *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **1032** (2022). 166597.
33. International Atomic Energy Agency, Implementing Guide: Radiological Crime Scene Management, Nuclear Security Series No. 22-G, IAEA, Vienna (2014).