A New Matrix Correction Method for Nuclear Material Mass Measurement in Shuffler Neutron Interrogation Systems

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Date: 2024-06-27T00:00:00+00:00

Abstract

The presence of matrix materials in waste drums constrains the accuracy of active interrogation systems for nuclear material mass measurements, making it essential to ensure that nuclear safeguards measurement results are unaffected by matrix materials. Therefore, based on flux monitor responses, this study develops a novel matrix correction method building upon traditional correction approaches. To validate the effectiveness of the proposed method, a WM3210 PAN Shuffler system model and common matrix materials were constructed using the Geant4 toolkit, and simulation studies were performed. The results indicate that for U3O8 materials of varying enrichments and distribution states, both the traditional and novel correction methods can effectively mitigate the impact of matrix materials on nuclear material mass measurements; however, the new method exhibits superior correction capability compared to the traditional approach. For scenarios where U3O8 material is positioned at the matrix center, the new correction method yields an average relative deviation of 13.6% for the 235U mass, whereas for scenarios with U3O8 material uniformly dispersed throughout the matrix, the average relative deviation for the 235U mass is 7.78%.

Full Text

Matrix Correction for Nuclear Material Mass Measurement by Shuffler Neutron Interrogation System

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Abstract

[**Background**] The presence of matrix materials in waste drums limits the accuracy of special nuclear material (SNM) mass measurements by active interrogation systems, and ensuring that measurement results for nuclear safeguards are unaffected by matrix materials is crucial. [**Purpose**] Therefore, considering the variation of delayed neutron count rate with increasing SNM mass, this paper develops a new matrix correction method based on traditional correction methods. [**Methods**] To verify the effectiveness of the new method, a WM3210 PAN Shuffler system model was constructed using the Geant4 toolkit, and correction algorithm research was conducted for common matrix materials. [**Results**] The results show that for U3O8 materials with different enrichments and distribution states, both traditional and new correction methods can effectively reduce the influence of matrix materials on SNM mass measurement, but the new method demonstrates better correction capability. For U3O8 material located at the matrix center, the new method yields an average relative deviation of 13.6% for 235U mass, while for U3O8 material uniformly dispersed in the matrix, the average relative deviation is 7.78%. [**Conclusions**] These results confirm the effective performance and practicality of the new correction method.

Keywords: Nuclear safeguard, Delayed neutron, matrix effect, Monte Carlo simulation

1 Measurement Principle and Model Construction

Shuffler System Modeling

The WM3210 PAN Shuffler system is a nuclear safeguards instrument based on delayed neutrons, commonly used for mass measurement of SNMs in 208 L waste drums [14]. The Shuffler system comprises a detection system and corresponding electronics and data processing modules. The detection system features an octagonal horizontal cross-section and is divided into upper and lower sections. The upper section includes the interrogation source shielding assembly, interrogation source storage assembly, and servo motor drive system, while the lower section constitutes the counting chamber for placing the waste drum under measurement. Six detector arrays are arranged on the side walls of the counting chamber, with each array consisting of seven 3He proportional counter tubes and HDPE shielding. Additionally, one detector array is installed at both the top and bottom of the counting chamber, each comprising twelve 3He proportional counter tubes and HDPE shielding. These 3He proportional counter tubes share identical physical properties, including an internal pressure of 10 atm, a diameter of 2.54 cm, and a length of 99.06 cm. Furthermore, the counting chamber is lined with 0.762 mm-thick metallic cadmium (Cd) to absorb thermal neutrons returning to the counting chamber (fast mode). The detection system model constructed using the Geant4 toolkit is shown in Figure 1 [Figure 1: see original paper]; for model simplification, the mechanical structures of the upper detection system were omitted.

Figure 1. Simulation model of WM3210 PAN Shuffler system, in which, (a) is side view, (b) is top view

The Shuffler system employs a 252Cf neutron source with an intensity of 1.23×10^{9} n · s^{-1} as the interrogation source. When the system is inactive or in passive measurement mode, the interrogation source is stored in the source storage assembly. In active interrogation mode, the system operational sequence is illustrated in Figure 2 [Figure 2: see original paper], comprising three fundamental steps: (a) background neutron counting measurement, (b) the servo motor transfers the interrogation source from the source storage chamber to the counting chamber via a transfer tube to irradiate the waste drum, and (c) after completing irradiation, the interrogation source is returned to the storage chamber via the transfer tube, followed by delayed neutron counting. Due to the low yield of delayed neutrons, steps (b) and (c) are typically repeated multiple times during a complete measurement to accumulate sufficient delayed neutron counts.

Figure 2. Schematic representation of the WM3210 PAN Shuffler system

The QGSP_{{{BIC}}*{{HP}}} physics list provided in the Geant4 toolkit encompasses various nuclear interaction processes and physical models, commonly employed for neutron-related physics simulations. However, the neutron elastic scattering function in the QGSP*{{{BIC}}*{{HP}}} physics list lacks descriptions of scattering processes for neutrons with energies below 4.0 eV. Considering that this work involves neutron moderation processes in multiple materials, we supplemented the QGSP*{{{BIC}}_{{HP}}} physics list with additional scattering processes for neutrons below 4.0 eV, incorporating preset neutron elastic scattering functions. Furthermore, the current Geant4 toolkit (Version 11.03) does not directly provide information related to delayed neutrons. Therefore, an algorithm was developed based on the Geant4 toolkit to simulate delayed neutron production and associated physical processes. This algorithm records, filters, and samples fragment information from neutron-induced fission of nuclear materials to identify precursor nuclei capable of producing delayed neutrons and generates new source terms. By reading the source term information into the Geant4 toolkit, simulation of delayed neutron production and related physical processes can be realized. All data processing in this work was completed using the Root data analysis framework [15].

Shuffler System Performance Verification

To verify the reliability of the Shuffler system model and simulation methodology, this work utilized the Geant4 toolkit to construct 252Cf and delayed neutron source terms to determine the detection efficiency of the Shuffler system model. Additionally, two U3O8 samples (designated UM1 and UM2) were constructed based on the Geant4 toolkit to calculate the proportion of delayed neutrons in 235U-induced fission events and the delayed neutron count rate per unit mass of 235U. Detailed parameters of the samples are listed in Table 1 .

Table 1. Standard sample physical properties

The main performance parameters of the system model are presented in Table 2 . The simulation results indicate that the detection efficiencies for 252Cf neutrons and delayed neutrons are 21.5% and 26.3%, respectively, with deviations of 10.3% and 1.5% from the Shuffler system reference values. In simulated measurements of the UM1 sample, delayed neutrons accounted for 1.57% of total fission neutrons, closely approaching the reference value of 1.60%. The delayed neutron count rate per unit mass of 235U calculated through the UM2 sample was 2.70 c·(s·g)^-1, slightly higher than the reference value by 3.85%. Considering errors introduced by model construction and the delayed neutron sampling algorithm, these results fully validate the accuracy of the Shuffler system model and the reliability of the simulation methodology.

Table 2. The main performance of the system model

2.1 Matrix Effects on Counting

Hydrogenous materials are among the most effective neutron moderators, and their density in matrix materials is commonly used to evaluate matrix moderating capability [19]. This work utilized the Geant4 toolkit to construct a limited but representative set of matrix materials and three groups of U3O8 materials to analyze matrix effects on delayed neutron count rates. Table 3 lists detailed characteristics of these matrices, including material density ( ) and hydrogen density (H) parameters. The physical dimensions of U3O8 materials reference the NBS-SRM-969 standard nuclear material, with masses ranging from 5 g to 600 g and positioned at the matrix center. Considering the characteristics of SNMs in spent fuel reprocessing plants, the 235U enrichment in U3O8 materials was set at 0.7%, 2%, and 3%, respectively.

Table 3. Matrix material physical properties

Table 4 summarizes the simulated results of delayed neutron count rates per unit uranium mass for U3O8 materials, where the delayed neutron count rate per unit uranium mass represents the average across different U3O8 material masses. Observation reveals that as H in matrix materials increases, the delayed neutron count rates for U3O8 materials with different 235U enrichments exhibit a trend of initial increase followed by decrease. Appropriate H in matrix materials can reduce interrogating neutron energy, thereby increasing the reaction cross-section with 235U in U3O8 to generate more fission events. However, as H further increases, excessive H leads to competition between neutron moderation and induced fission, consequently reducing the number of induced fission events. Therefore, the influence of matrices with different H on delayed neutron count rates is complex and cannot be simply generalized.

Table 4. Delayed neutron count rate per unit uranium mass for the U3O8 material at different 235U enrichment and matrix material

2.2 Principle of Matrix Effect Correction

If matrix material types are known a priori, dedicated calibration curves can be constructed in practical applications to compensate for matrix effects on measurements, thus obviating the need for matrix effect correction. However, in actual safeguards measurements, detailed information about matrix materials is often unavailable. Therefore, the primary objective of developing matrix effect correction methods is to compensate for effects caused by unknown matrix materials on delayed neutron counts to obtain true delayed neutron signals.

Based on the characteristic that the presence of matrix materials affects neutron flux and energy distribution inside the counting chamber, Rinard et al. proposed a matrix effect correction method utilizing neutron flux monitor responses. The neutron flux monitor consists of two 3He proportional counter tubes positioned on the counting chamber side walls. These two 3He proportional counter tubes share identical diameter, length, and pressure (2.54 cm, 99.06 cm, and 4 atm, respectively), with the sole distinction being that one is coated with metallic cadmium (Cd tube) while the other is uncoated (bare tube). During the Shuffler process, interactions with matrix materials cause interrogating neutrons to scatter out of the waste drum and be recorded by the flux monitor. Since metallic cadmium possesses a large absorption cross-section for thermal neutrons, the Cd tube exhibits greater sensitivity to fast neutrons within the counting chamber, whereas the bare tube demonstrates higher response to thermal neutrons.

The ratio of neutron counts between the bare tube and Cd tube is defined as R, termed the matrix effect coefficient. R is independent of interrogation source intensity and depends solely on the degree of moderation of interrogating neutrons by matrix materials. Consequently, R can be used to quantify variations in neutron flux within the counting chamber. The scenario of a waste drum without any matrix material filling is defined as zero matrix; under ideal conditions (neglecting the influence of waste drum wall materials on interrogating neutrons), the R value for a zero-matrix waste drum should approach 1. This enables establishment of a multi-parameter analysis model incorporating R, delayed neutron count rate, and 235U mass, achieving correction of delayed neutron counts by exploiting the differential response of flux monitors to fast and thermal neutrons.

2.3 Effect Correction Method

The correction method based on flux monitor response was first proposed by Rinard et al. For the Shuffler system, fast neutrons released by 252Cf can effectively penetrate SNM of certain mass. Therefore, in zero matrix conditions, the relationship between delayed neutron count rate and 235U mass can be considered linear,

$$d = K \cdot m$$

where d represents the true delayed neutron count rate, m denotes the mass of 235U in the U3O8 material, and K is the delayed neutron count rate per unit mass of 235U, also known as the mass calibration coefficient.

When matrix materials are present, d changes due to interactions between neutrons and the matrix. Let the delayed neutron count rate under these conditions be md; the relationship between d and md can be described by the following equation,

$$m_d = d \cdot f(R) = K \cdot m \cdot f(R)$$

where f(R) is the matrix effect correction function, f(R) = p__1 R^{p__2} is a function related to R, and p__1, p__2 are fitting parameters.

By pre-measuring a series of different matrix materials, corresponding md and R values can be obtained. Using Equation (2) to derive f(R) for each matrix, the fitting relationship between f(R) and R can be constructed, ultimately yielding the parameters p__1 and p__2.

However, for SNM objects spanning a certain mass range, the relationship between delayed neutron count rate and 235U mass is not absolutely linear, meaning K is not constant. As shown in Figure 3 [Figure 3: see original paper], it can be observed that as the mass m of 235U in U3O8 material increases, the K value generally exhibits a gradually decreasing trend. This occurs because increasing SNM mass leads to volume changes, preventing interrogating neutrons from reaching all fissile nuclei within the sample with equal probability. Consequently, non-uniform induced fission probabilities arise within the SNM sample volume, and the presence of matrix materials further reduces interrogating neutron energy, exacerbating this effect.

Figure 3. For the 93.2%-enriched U3O8 material, the trend of the unit 235U mass delayed neutron count rate (K) with the change of 235U material mass (m), the dashed line represents the fitted curve using the logarithmic function.

To better meet the requirements of nuclear safeguards, this paper proposes a new matrix correction method based on the traditional approach, incorporating the influence of SNM self-shielding of interrogating neutrons into the correction methodology. This method employs Equation (4) to describe the true relationship between d and mass m,

$$d = K(m) \cdot m = (p_3 \cdot \ln(m) + p_4) \cdot m$$

where p__3 and p__4 are fitting parameters.

Combining with Equation (2) yields:

$$m_d = K(m) \cdot m \cdot f(R) = (p_3 \cdot \ln(m) + p_4) \cdot m \cdot p_1 \cdot R^{p_2}$$

Rearranging gives:

$$m = \frac{m_d}{p_1 \cdot R^{p_2} \cdot (p_3 \cdot \ln(m) + p_4)}$$

The four parameters in Equation (6) can be determined by simulating md and R for U3O8 materials of different masses in various matrix materials, thereby incorporating the influence of neutron self-shielding effects during SNM interrogation into the correction method. By substituting the determined parameters along with md and R measured from unknown matrix materials into the equation, the mass of 235U in unknown matrices can be solved.

3 Matrix Effect Correction Results

To verify the correction effectiveness of the new method on matrix effects, this work constructed multiple groups of U3O8 materials with masses ranging from 1 g to 1000 g, with 235U enrichments set at 0.7%, 2%, and 3%, respectively. Furthermore, these U3O8 materials exhibited two different distribution states: as a solid block located at the matrix center and as uniformly dispersed throughout the matrix.

3.1 Nuclear Material at Matrix Center

U3O8 material located at the matrix center is considered an extreme measurement scenario. By sequentially filling the matrix materials from Table 3 into the waste drum, the corresponding fitting parameter values in Equations (3) and (6) can be obtained. Using these two correction methods to correct several matrix materials with different R values, the relative deviation between corrected and true 235U mass can be compared to evaluate matrix effect correction performance.

The results are presented in Figure 4 [Figure 4: see original paper], where RD_{raw} represents the average relative deviation of 235U mass without correction, RD_{tcor} denotes the traditional method, and RD_{ncor} represents the new correction method. For U3O8 materials with different 235U enrichments, the relative deviation of 235U mass ranges from 34.2% to 690% without correction. After correction using the traditional method, the average relative deviations of 235U mass for U3O8 materials with enrichments of 0.7%, 2%, and 3% are 22.3%, 24.3%, and 24.7%, respectively, whereas the new method yields average relative deviations of 10.9%, 14.3%, and 15.7%, respectively. These results effectively demonstrate that the new method, which accounts for SNM self-shielding of interrogating neutrons, possesses superior correction capability for matrix effects.

Figure 4. The average relative deviation in mass of 235U when U3O8 material located at the center of the matrix, in which, (a) is 0.7%-enriched U3O8 material, (b) is 2%-enriched U3O8 material, (c) is 3%-enriched U3O8 material.

3.2 Nuclear Material Uniformly Dispersed in Matrix

U3O8 material uniformly dispersed in the matrix can also be considered another extreme measurement scenario, as uniformly dispersed U3O8 leads to significant fluctuations in delayed neutron count rates. Using the same method to correct several matrix materials with different R values, the results are shown in Figure 5 [Figure 5: see original paper]. For U3O8 materials with enrichments of 0.7%, 2%, and 3%, the average relative deviations of 235U mass calculated by the traditional method are 26.1%, 22.7%, and 17.3%, respectively. In contrast, the new correction method demonstrates better matrix effect correction performance, yielding average relative deviations of 7.29%, 9.71%, and 6.33%, respectively. These results indicate that the developed new correction method achieves superior correction effectiveness.

Figure 5. The average relative deviation in mass of 235U when U3O8 material homogeneous dispersed throughout the matrix, in which, (a) is 0.7%-enriched U3O8 material, (b) is 2%-enriched U3O8 material, (c) is 3%-enriched U3O8 material.

Based on the influence of matrix materials on neutron flux and energy within the counting chamber, Rinard et al. proposed a matrix effect correction method utilizing neutron flux monitors. However, Rinard's method did not consider the potential impact of SNM self-shielding of interrogating neutrons on correction effectiveness. This paper introduces consideration of interrogating neutron self-shielding effects based on Rinard's work, thereby developing a new correction method. To analyze the influence of matrix materials on delayed neutron counting and specifically validate the rationality of the improved method, this work constructed a WM3210 PAN Shuffler system model and a series of matrix materials using the Geant4 toolkit, and conducted simulation validation studies.

Simulation results demonstrate that appropriately moderating interrogating neutron energy can increase its induced fission reaction cross-section with SNM; thus, low-H matrices can significantly increase delayed neutron count rates. However, for matrices with strong moderating capability, excessive moderation of both interrogating and delayed neutrons causes a substantial decrease in detected delayed neutron count rates. Correction results for multiple matrix materials with different R values indicate that both traditional and new correction methods can achieve good correction performance. Using average relative deviation of 235U mass as the evaluation criterion, the new correction method yields smaller relative deviations in 235U mass compared to the traditional method. When located at the matrix center, the new method produces average relative deviations of 10.9%, 14.3%, and 15.7% for U3O8 materials with enrichments of 0.7%, 2%, and 3%, respectively; when uniformly dispersed in the matrix, the new method yields average relative deviations of 7.29%, 9.71%, and 6.33%, respectively.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (No. 12227810).

Author Contributions

Yong Jinlong was responsible for simulation, data processing, compiling writing materials, and drafting the manuscript; Song Yushou provided the research concept, reviewed and ensured scientific rigor, and coordinated manuscript revisions; Zhao Yunlong conducted preliminary investigations, simulations, and technical summaries; Hou Yingwei provided simulation guidance and researched correction methods; Liu Huilan was responsible for data collection and organization.

References

1. Tsuchiya H, Kitatani F, Maeda Y, et al. Development of neutron resonance transmission analysis as a non-destructive assay technique for nuclear nonproliferation[J]. Plasma and Fusion Research, 2018, 13(1): 2406004. DOI: 10.1585/pfr.13.2406004.
2. Menlove H O. Passive neutron waste-drum assay with improved accuracy and sensitivity for plutonium using the add-a-source method[R]. Institute of Nuclear Materials Management, 1992.
3. Seabury E H, Chichester D L, Wharton, C J, et al. A comparison of neutron-based non-destructive assessment methods for chemical warfare materiel and high explosives[J]. In AIP Conference Proceedings, 2009, 1099: 928-931. DOI: 10.1063/1.3120193.
4. Remetti R, Gandolfo G, Lepore L, et al. In field application of differential Die-Away time technique for detecting gram quantities of fissile materials[J]. Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2017, 870: 123-130. DOI: 10.1016/j.nima.2017.07.033.
5. Ogren K, Wu T, Nattress J, et al. The effects of low-Z shielding on uranium isotope discrimination using the time-emission profiles of long-lived delayed neutrons[J]. Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2021, 1019: 165847. DOI: 10.1016/j.nima.2021.165847.
6. Caldwel J T, Hastings R D, Herrera G C, et al. The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers[R]. Los Alamos National Laboratory, 1986.
7. Simon E, Jallu F, Pérot B, et al. Feasibility study of fissile mass quantification by photofission delayed gamma rays in radioactive waste packages using MCNPX[J]. Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 2016, 840: 28-35. DOI: 10.1016/j.nima.2016.09.047.

8. Komeda M, Toh Y. Conceptual study on a novel method for detecting nuclear material using a neutron source[J] Annals Nuclear Energy, 2020, 135: 106993. DOI: 10.1016/j.anucene.2019.106993.
9. Jallu F, Loche F. Improvement of non-destructive fissile mass assays in α low-level waste drums: A matrix correction method based on neutron capture gamma-rays and a neutron generator[J]. Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms, 2018, 266(16): 3674-3690. DOI: 10.1016/j.nimb.2008.06.013.
10. Rinard P M, Adams E L, Menlove H O, et al. The NondestructiveAssay of 55-Gallon Drums Containing Uranium and Transuranic Waste Using Passive-Active Shufflers[R]. Los Alamos National Laboratory, 1993.
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. DOI: 10.1016/S0168-9002(03)01368-8.
12. Allison J, Amako K, Apostolakis J, et al. Geant4 Developments and Applications[J]. IEEE Transactions on Nuclear Science, 2006, 53(1): 270- 278. DOI: 10.1109/TNS.2006.869826.
13. Allison J, Anako 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, 835(1): 186-255. DOI: 0.1016/j.nima.2016.06.125.
14. Lin W, Li D H, Li Z P, et al. Application and prospect of the shuffler drum waste measurement device in nuclear material accounting[J]. Instrumentation Users, 2023, 30(5): 45-49. DOI: 10.3969/j.issn.1671-1041.2023.05.010.
15. Brun R, Rademakers F, ROOT - An object oriented data analysis framework[J]. Nuclear Instruments and Methods in Physics Research, Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 1997, 389(1-2): 81-86. DOI: 10.1016/S0168-9002(97)00048-X.
16. Menaa N, McElroy R D, Kane Smith S, Kirkpatrick J, et al. Cf-252 Shuffler for Waste Assays and Safeguards Applications[C]. 51th Annual Meeting of the Institute of Nuclear Materials Management, Baltimore, USA, 2010.
17. Rinard P M. Application Guide to Shufflers[R]. Los Alamos National Laboratory, 2001.

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