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

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Abstract

The presence of matrix materials in waste drums limits the accuracy of active interrogation systems for nuclear material mass measurement, making it crucial to ensure that nuclear safeguards measurement results are unaffected by matrix materials. Therefore, this paper develops a novel matrix correction method based on flux monitor response, building upon traditional correction approaches. To validate the effectiveness of the proposed method, the Geant4 toolkit was employed to construct a model of the WM3210 PAN Shuffler system along with common matrix materials. The results demonstrate that for U3O8 materials with varying enrichment levels and distribution states, both traditional and new correction methods can effectively mitigate the influence of matrix materials on nuclear material mass measurements. However, the new method exhibits superior correction capability compared to the traditional approach. Specifically, for U3O8 material positioned at the matrix center, the new correction method yields an average relative deviation of 13.6% for 235U mass, whereas for U3O8 material uniformly dispersed throughout the matrix, the average relative deviation is 7.78%.

Full Text

Preamble

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

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Abstract

[**Background**] Active neutron interrogation (ANI) measurement systems can quantify the fissile mass of special nuclear materials (SNMs) using neutrons and are widely used in nuclear safeguard fields. However, the hydrogen-containing matrix in waste drums reduces the signal of delayed neutrons and limits the fissile mass measurement precision of the ANI system. [**Purpose**] It is essential to ensure that the fissile mass evaluation of the SNM is independent of matrix material and to improve the assay performance of the ANI system in nuclear safeguard measurements. [**Methods**] Therefore, based on the flux monitor response, a new matrix correction method was developed on the basis of traditional correction methods in this work. [**Results**] It turned out that the effect of various hydrogen densities on the delayed neutron count rate differs. For U3O8 materials with different enrichment and distribution, matrix effect may result in an overestimation of up to ~7 times in the observed mass of 235U. For U3O8 materials with different 235U enrichments and distribution states, both the traditional correction method and the new correction method can effectively reduce the influence of the matrix material on the measurement of SNM mass. But compared with the traditional method, the new method can achieve better correction results. For the case where U3O8 material is located in the center of the matrix, the average relative deviation of 235U mass obtained by the new correction method is 13.6%, while for the case where U3O8 material homogeneous dispersed throughout the matrix, the average relative deviation of 235U mass obtained by the new correction method 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

Introduction

Rapid and robust verification of special nuclear materials (SNMs) within nuclear industrial facilities is crucial for nuclear material management and safeguards. In facilities such as spent fuel reprocessing plants, SNMs in nuclear waste typically exist in the form of oxides or mixtures. Waste drums storing SNMs are also filled with materials such as iron filings, polyethylene, and fiberboard. Additionally, in cases of illicit trafficking of SNMs, hiding SNMs in special containers to deceive inspection detectors is a common practice. These materials disguise SNMs by altering their intrinsic characteristic signals. In nuclear safeguards and monitoring, substances in the measured object other than the SNM of interest are referred to as matrix materials.

For active neutron interrogation, the moderating properties of matrix materials not only change the interrogating neutron flux and energy distribution but also affect secondary product neutrons released from SNM-induced fission. Consequently, the number of neutron signals reaching the detector varies with different matrix material types. Although current active interrogation techniques have proven extremely sensitive to the presence of SNMs, the primary objective of safeguards measurements remains accurate acquisition of SNM mass information. The presence of matrix materials complicates the practical application of active interrogation techniques. In nuclear safeguards measurements, providing precise SNM mass information and ensuring that mass measurement results are independent of matrix materials represents a technical challenge. To meet the relevant needs of nuclear safeguards, appropriate methods must be applied to compensate for SNM mass measurement biases caused by matrix materials, thereby improving the accuracy of nuclear safeguards measurements.

To address these challenges, Rinard et al. at Los Alamos National Laboratory (LANL) developed a matrix effect correction method based on the response differences of 3He proportional counters to interrogating neutron flux and energy. However, this method neglects the shielding effect of the SNM itself on interrogating neutrons, resulting in biases that cannot be ignored in practical measurements. Therefore, this paper proposes a new correction method to improve correction effects for different matrices. To verify the applicability of the new method, the Geant4 toolkit was used to construct a WM3210 Passive/Active Neutron (PAN) Shuffler measurement system model and various common matrix materials, allowing validation of the new correction method's effectiveness for different matrix materials and comparison with traditional methods.

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. The Shuffler system comprises a detection system and corresponding electronics and data processing modules. The detection system has 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 is the counting cavity for placing the waste drum to be measured. Detector groups are arranged on all six side walls of the counting cavity, with each group consisting of 7 3He proportional counters and HDPE shielding. Additionally, one detector group is arranged at the top and bottom of the counting cavity respectively, each consisting of 12 3He proportional counters and HDPE shielding. These 3He proportional counters have identical physical properties, including 10 atm internal pressure, 2.54 cm diameter, and 99.06 cm length. Furthermore, the counting cavity is lined with 0.762 mm thick metallic cadmium (Cd) to absorb thermal neutrons returning to the counting cavity (fast mode). The detection system model constructed based on the Geant4 toolkit is shown in Figure 1 [Figure 1: see original paper]. For model simplification, the mechanical structure of the upper section of the detection system was ignored.

The Shuffler system uses a 252Cf neutron source with an intensity of 1.23×10^{9} n/s as the interrogation source. When the system is not operating or in passive measurement mode, the interrogation source is stored in the source storage assembly. In active interrogation mode, the system operation process is shown in Figure 2 [Figure 2: see original paper], which includes three basic steps: (a) background neutron count measurement, (b) the servo motor transfers the interrogation source from the source storage chamber to the counting cavity 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) typically need to be repeated multiple times in a complete measurement to accumulate sufficient delayed neutron counts.

The QGSP_{{{BIC}}*{{HP}}} physics list provided in the Geant4 toolkit includes various nuclear interaction processes and physical models. Therefore, based on the QGSP*{{{BIC}}_{{HP}}} physics list, this work further added neutron thermal scattering processes to more accurately simulate neutron moderation and diffusion processes in the system. Additionally, the current Geant4 toolkit (Version 11.03) cannot directly provide delayed neutron-related information. Therefore, an algorithm was developed based on the Geant4 toolkit to simulate delayed neutron generation and its related physical processes. All data were processed using the Root data analysis framework.

Shuffler System Performance Verification

The main performance characteristics of the Shuffler system model are shown in Table 1 . The model's 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. Simulation results for U3O8 material UM1 (whose physical details are shown in Table 1) indicate that delayed neutron signals account for only 1.57% of the total fission neutron signals, which is close to the reference value of 1.60%. Material UM2 consists of a series of U3O8 materials with masses ranging from 1.25-250.21 g to measure the delayed neutron count rate per unit mass of 235U. Simulation results show that the simulated delayed neutron count rate per unit mass of 235U is 2.70 $c\cdot(s\cdot g)^{-1}$, slightly 3.85% higher than the reference value. Considering errors from model construction and the delayed neutron sampling algorithm, these results effectively verify the reliability of the Shuffler system model.

2 Matrix Effect Analysis and Correction Method Development

2.1 Influence of Matrix on Counting Rate

Hydrogen-containing materials are among the most effective neutron moderators, and their density in matrix materials is typically used to evaluate the matrix moderating capability. This work used the Geant4 toolkit to construct a limited but representative set of matrix materials and three groups of U3O8 materials to analyze the influence of matrix on delayed neutron counting rate. Table 3 lists the detailed characteristics of these matrices, such as material density ( ) and hydrogen density (_H). The physical dimensions of U3O8 materials reference the NBS-SRM-969 standard nuclear material, with masses of 5-600 g and located 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 4 summarizes the changes in delayed neutron counting rate per unit uranium mass for U3O8 materials, where the counting rate per unit uranium mass is the average value across different U3O8 masses. Observation reveals that as _H in the matrix material increases, the delayed neutron counting rate for U3O8 materials with different 235U enrichments first increases and then decreases. Appropriate _H in matrix materials can reduce interrogating neutron energy, thereby increasing the reaction cross-section with 235U in U3O8 to produce more fission events. However, as _H further increases, excessive hydrogen density leads to competition between neutron moderation and induced fission, 反而 reducing the number of induced fission events. Therefore, the influence of matrices with different _H on delayed neutron counting rates is complex and cannot be simply generalized.

2.2 Principle of Matrix Effect Correction

If the matrix material type is known a priori, dedicated calibration curves can be constructed in practical applications to compensate for matrix effects on measurements, thus eliminating the need for matrix effect correction. However, in actual safeguards measurements, detailed information about matrix materials is often lacking. Therefore, the main purpose of developing matrix effect correction methods is to compensate for the influence of unknown matrix materials on delayed neutron counting to obtain the true delayed neutron signal.

Based on the characteristic that matrix materials affect the flux and energy distribution of neutrons inside the counting cavity, Rinard et al. proposed a matrix effect correction method based on neutron flux monitor response. The neutron flux monitor consists of two 3He proportional counters placed on the side wall of the counting cavity. These two 3He proportional counters have identical diameter, length, and pressure (2.54 cm, 99.06 cm, and 4 atm, respectively). The only difference is that one is wrapped with metallic cadmium (Cd tube) while the other is not (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 has a large absorption cross-section for thermal neutrons, the Cd tube is more sensitive to fast neutrons in the counting cavity. In contrast, the bare tube has higher response to thermal neutrons.

The ratio of neutron counts between the Cd tube and bare tube is defined as R, called the matrix effect coefficient. R is independent of interrogation source intensity and depends only on the degree of moderation of interrogating neutrons by matrix materials. Therefore, R can be used to quantify changes in neutron flux within the counting cavity. The case where the waste drum contains no matrix material is defined as zero matrix. Under ideal conditions (ignoring the influence of waste drum material on interrogating neutrons), the R value for a zero-matrix waste drum should be close to 1. Thus, a multi-parameter analysis model including R, delayed neutron counting rate, and 235U mass can be established to achieve correction of delayed neutron counting using the response differences of flux monitors to fast and thermal neutrons.

The R values and flux monitor counting rates for various matrices calculated using the Geant4 toolkit are shown in Table 5 . The simulated R values initially increase and then decrease with increasing _H, showing a similar trend to delayed neutron counting rates. The bare tube is more sensitive to changes in _H within the matrix, with more significant fluctuations in counting rate compared to the Cd tube.

2.3 Development of Correction Method

The correction method based on flux monitor response was first proposed by Rinard et al. It assumes a linear relationship between delayed neutron counting rate and 235U mass at zero matrix, expressed by Equation (1):

$$d = K \cdot m$$

where d represents the true delayed neutron counting rate, m represents the mass of 235U in U3O8 material, and K is the delayed neutron counting rate per unit mass of 235U, also called the mass calibration constant.

When matrix materials are present, the interaction between matrix and neutrons causes d to vary with different matrix materials, which can be represented by R. Therefore, the relationship between md (actual delayed neutron counting rate) and d can be described by Equations (2) and (3):

$$m_d = d \cdot p(R)$$

$$p(R) = \sum_{j=0}^{n} a_j R^j$$

where p(R) is the matrix effect correction function related to R, and a_j are fitting parameters. Thus, by pre-measuring a series of different matrix materials, the corresponding md and R can be obtained. Through Equation (2), p(R) for the corresponding matrix can be obtained, and by constructing the fitting relationship between p(R) and R, the parameters can be determined.

However, for SNM objects with a certain mass range, the relationship between delayed neutron counting rate and 235U mass is not absolutely linear, meaning K is not constant. As shown in Figure 4 [Figure 4: see original paper], it can be observed that as the mass m of 235U in U3O8 material increases, the K value tends to gradually decrease. This is because as SNM mass increases, its volume changes, preventing interrogating neutrons from reaching all fissile nuclei within the sample with equal probability, resulting in non-uniform induced fission probabilities within the SNM sample volume. The presence of matrix materials further reduces interrogating neutron energy, exacerbating this effect.

Therefore, to better meet nuclear safeguards requirements, this paper proposes a new matrix correction method based on traditional methods, incorporating the influence of SNM interrogating neutron self-shielding into the correction method. This method uses Equation (4) to represent the true relationship between d and m:

$$d = \frac{m}{b_1 + b_2 \ln(m) + b_3}$$

where b_1, b_2, and b_3 are fitting parameters. From Equation (2), we can derive:

$$m_d = \frac{m}{b_1 + b_2 \ln(m) + b_3} \cdot p(R)$$

Rearranging the above equation, we can further derive Equation (6):

$$m = m_d \cdot \frac{b_1 + b_2 \ln(m) + b_3}{p(R)}$$

The four parameters in Equation (6) can be determined by simulating md and R for U3O8 materials of different masses in different matrix materials. Thus, it incorporates the influence of SNM interrogating neutron self-shielding effect on correction results into the correction method. By using the determined parameters along with md and R obtained from measuring unknown matrix materials, the mass of 235U in unknown matrices can be solved.

3 Matrix Effect Correction Results

To verify the correction effect of the new method on matrix effects, multiple groups of U3O8 materials with masses of 1-1000 g were constructed in this work, with 235U enrichments set at 0.7%, 2%, and 3%, respectively. Additionally, these U3O8 materials had two different distribution states: located as a block at the center of the matrix and homogeneously dispersed throughout the matrix.

3.1 Nuclear Material Located 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 matrix effect correction performance can be evaluated by comparing the average relative deviation between the corrected 235U mass and the true 235U mass.

As shown in Figure 5 [Figure 5: see original paper], rawRD represents the average relative deviation of 235U mass without correction, RD_t-cor represents the traditional correction method, and RD_n-cor represents the new correction method. For U3O8 materials with different 235U enrichments, the relative deviation of 235U mass without correction ranges from 34.2% to 690%. 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. In contrast, the average relative deviations obtained using the new method are 10.9%, 14.3%, and 15.7%, respectively. These results effectively demonstrate the superior matrix effect correction capability of the new method that considers SNM interrogating neutron self-shielding effects.

3.2 Nuclear Material Homogeneously Dispersed in Matrix

U3O8 material homogeneously dispersed in the matrix can also be considered an extreme measurement scenario, as uniform dispersion of U3O8 will cause significant fluctuations in delayed neutron counting rates. Using the above two methods to correct several matrix materials with different R values, the results are shown in Figure 6 [Figure 6: 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 comparison, the new correction method demonstrates better matrix effect correction performance, yielding average relative deviations of 235U mass of 7.29%, 9.71%, and 6.33%, respectively.

Based on the influence of matrix materials on neutron flux and energy within the counting cavity, Rinard et al. proposed a matrix effect correction method using neutron flux monitors. However, Rinard's method did not consider the potential influence of SNM interrogating neutron self-shielding on correction performance. Therefore, 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 particularly to verify the rationality of the improved method, this work used the Geant4 toolkit to construct a WM3210 PAN Shuffler system model and a series of matrix materials.

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

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Author Contributions

YONG Jinlong was responsible for simulation, data processing, summarizing writing materials, and drafting the paper. SONG Yushou was responsible for providing ideas, reviewing and ensuring the scientific quality of the paper, and coordinating revisions. ZHAO Yunlong was responsible for preliminary research, simulation, and technical summary. HOU Yingwei was responsible for simulation guidance and correction method research. LIU Huilan was responsible for data collection and organization.

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