DOI: 10.15669/pnst.8.377

Progress in Nuclear Science and Technology
Volume 8 (2025) pp. 377-380

ARTICLE

Development of a Table-Top NRTA System: Monte Carlo Study of Collimator/Shielding for Pu Pellet Sample Measurement

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The present Monte Carlo study was conducted to evaluate the efficacy of a table-top Neutron Resonance Transmission Analysis (NRTA) system for measuring small Nuclear Material (NM) samples, such as plutonium (Pu) pellets, using a compact ^{252}Cf spontaneous fission neutron source. Time-of-flight spectra obtained from a 2 cm^3 cylindrical Pu pellet sample exhibited resonance dips when subjected to collimator/shielding made of polyethylene (PE) that contained 20 % of B_2O_3 (B_2O_3-PE) and 50 % of LiF (LiF-PE). In contrast, the TOF spectra obtained using plain PE and air exhibited no discernible resonance dips. These results demonstrate the capability of the designed table-top NRTA system to measure small NM samples efficiently when employing suitable collimator/shielding materials. Given the results associated with B_2O_3-PE, it is planned to be used in implementing the physical table-top NRTA system.

KEYWORDS: Monte Carlo, neutron resonance transmission, Pu, nuclear material, neutron, collimator, B_2O_3

I. Introduction

In the context of escalating nuclear proliferation, there is a pressing need for diverse analytical methods that can be deployed in a complementary manner to assay nuclear fuel and quantify fissile material inventories accurately. A major challenge is the non-destructive analysis of nuclear fuel assemblies without resorting to disassembly, sectioning, chopping, or active sampling while ensuring consistency with head-end inventory data for materials entering present and future generations of spent nuclear fuel. A nuclear technique that meets these criteria, while also providing precise quantitative measurements of plutonium (Pu) inputs to safeguarded nuclear fuel facilities before any material decomposition, could significantly enhance the reliability of current plutonium mass assessments, which predominantly depend on vendor-supplied burn-up calculations.[1,2)]

Neutron Resonance Transmission Analysis (NRTA) is a promising nondestructive assay (NDA) method for the qualitative and quantitative assessments of uranium (U), Pu, higher-order actinides, and fission products within spent fuel assemblies.[3)] Nevertheless, conventional NRTA systems usually require neutron generation through a complex, multi-step process involving large particle accelerators.[4-7)] To facilitate the downsizing of NRTA systems for improved mobility and in-situ applications, using spontaneous fission neutron sources, such as ^{252}Cf, could offer a viable alternative. This would necessitate employing appropriate collimators to shield background neutrons and shape the neutron flux.

This study uses Monte Carlo (MC) methods to evaluate the efficacy of nuclear material (NM) detection for small-scale samples, specifically Pu pellets, using the table-top NRTA system under development.

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II. Materials and Methods

1. Basic Principle of NRTA

To quantify NMs, the NRTA technique employs the time-of-flight (TOF) method. Neutrons emitted from a neutron source are directed toward the measurement sample located mid-way between the neutron source and the detector. Among these, neutrons that pass through the sample without undergoing scattering, absorption, or fission are captured by a downstream neutron detector. The detection time of these unscattered neutrons is recorded. By calculating the time difference between this detection time and the initiation time from the neutron source, we can determine the TOF of the neutrons and subsequently convert it to neutron energy, E_n, using the following equation[8,9)]:

$$E_n = \frac{1}{2} m_n (L/t_n)^2. \quad (1)$$

The variables m_n represent the neutron mass and L the flight path length, respectively. The number of transmitted neutrons can be theoretically expressed using the following formula[8)]:

$$T(E_n) = I(E_n)/I_0(E_n) = \exp\left(-\sum \sigma_k (E_n)\, n_k\right), \quad (2)$$

where k designates each nuclide, and σ_k and n_k denote the total cross-section and areal density, respectively. This relationship indicates that larger resonant features and higher material densities lead to greater neutron attenuation. This attenuation manifests as resonance dips in the neutron TOF spectrum. The required cross-section values can be taken from nuclear data libraries such as JENDL,[10-12)] allowing us to evaluate the material properties by analyzing these resonance dips within the neutron TOF spectrum.

2. Collimator Materials and Setup

The simulated system comprises a neutron source (^{252}Cf and high-density polyethylene – HDPE), collimators, and a neutron detector, with a flight path length of 52 cm.[13] In this system, neutrons originating from ^{252}Cf pass through the collimator to irradiate the measurement sample. The selection of collimator materials in the design/development of the table-top NRTA system is essential for optimizing the neutron flux of energy between 0.1 eV and 10 eV (hereafter referred to as slow neutrons) at the target area during the neutron transport analysis.[14] Effective direction and shaping of neutron beams necessitate the use of various materials and techniques to shape the neutron trajectories and their dispersion for NRTA purposes. This study evaluates the performance and appropriateness of several materials including:

(1) Air, which serves as a no-cost baseline reference material.

(2) Polyethylene (PE) with 20% boric oxide (B_2O_3-PE): B_2O_3 was incorporated to enhance the neutron absorption properties as ^{10}B has a high neutron capture cross-section. As a result, B_2O_3-PE serves as an effective moderator and shield.

(3) PE with 50% lithium fluoride (LiF-PE), LiF-PE also has high thermal neutron absorption cross-sections and does not emit gamma rays when it absorbs neutrons. Thus, it is also considered to be a candidate.

(4) Standard PE.

In this study, a 10^4 Bq ^{252}Cf spontaneous fission neutron source was used. The source emits neutrons isotropically with an energy spectrum ranging from a few keV to around 20 MeV. To slow down the fast neutrons and achieve a thermalized neutron flux suitable for TOF measurements, a 3cm thick PE moderator was placed adjacent to the ^{252}Cf source. A neutron detector was simulated in a downstream position relative to the system, positioned in a central axial alignment with the ^{252}Cf source and PE moderator, to estimate the neutron flux for TOF assessment.

III. Monte Carlo Modeling

The Particle and Heavy Ions Transport code System (PHITS) v3.33 was used for the computational analyses in this study.[15,16] **Figure 1** delineates the geometrical configuration of the table-top NRTA system along with various collimator materials assessed. A key advantage of MC codes like PHITS is their proficiency in accurately modeling neutron interactions within a medium, encompassing processes such as elastic and inelastic scattering, neutron capture, fission, and secondary particle production.[14] The simulations leverage nuclear cross-section data sourced from JENDL5.[10] For comparative analysis, simulations were conducted with and without including a 2 cm³ cylindrical plutonium pellet sample (1.8 cm thickness, 0.6 cm radius) positioned in the sample lodge. The elemental composition of the Pu sample in the fuel pellet is detailed in **Table 1**.

In this work, the PHITS MC code was used to define the ^{252}Cf source as an isotropic source based on the energy spectrum and multiplicity data by Verbeke et al. (2014), which account for both spontaneous fission neutrons and the subsequent gamma rays.[17] The neutron detector was modeled as a plastic scintillator with a 2-inch diameter and 2-inch length, positioned downstream.

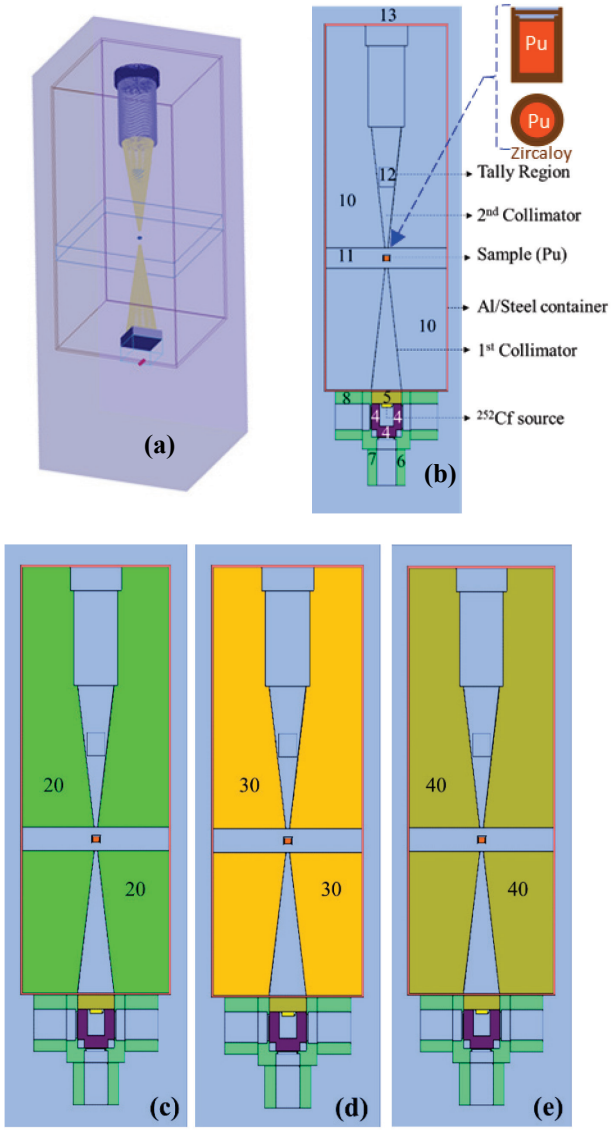

Fig. 1 (a) Simulated 3-D geometries of the table-top NRTA modeling under development (some parts were hidden). (b): 2-D geometry of the system under investigation with various configurations and no collimator material (air-filled space); (c) B_2O_3-PE; (d) LiF-PE; (e) PE. 4- LiF plate for gamma-ray signal filtration; 5- PE block (3 cm thick for neutron moderation); 6-, 7-, 8-, 9- Pb blocks for background gamma-ray shielding; 11- sample lodge; 12- tally region (EJ270 plastic scintillator); 13- exterior air to the system. 10- air 20- B_2O_3-PE; 30- LiF-PE; and 40- PE configurations, respectively

Table 1 Pu fuel pellet elemental composition and its density used for the simulation

Pu sample	Atomic fraction	Pu sample	Atomic fraction
^{16}O	7.44×10^{-3}	^{239}Pu	9.83×10^{-1}
^{24}Mg	2.04×10^{-1}	^{240}Pu	4.19×10^{-1}
^{25}Mg	2.73×10^{-2}	^{241}Pu	1.42×10^{-1}
^{26}Mg	3.17×10^{-2}	^{242}Pu	9.49×10^{-2}
^{238}Pu	2.79×10^{-2}	Density	1.162 g/cm^3

IV. Results and Discussion

Figure 2 presents the TOF spectra for the Pu pellet sample, both sample-in and sample-out spectra for neutron detection and **Fig. 3** for gamma-ray. In Fig. 2 (a), the neutron TOF spectra for various configurations (b to e from Fig. 1) are depicted. Notably, the TOF spectra for the Air and plain PE collimator configurations lack resonance dips, indicating their unsuitability for the table-top NRTA system. In contrast, the B_2O_3-PE and LiF-PE configurations distinctly show resonance dips associated with Pu. However, the LiF-PE configuration displays certain missing peaks, highlighting a deviation. These findings confirm that the table-top NRTA system, for which the collimators were evaluated using MC simulations, can effectively detect Pu isotopes in small sample sizes. This is particularly evident in configurations using B_2O_3-PE, which enhances detection performance.

For gamma-ray measurements, a different pattern is observed for both the Pu sample in and out spectra (Fig. 3). Beyond 15 μsec, the spectra dominated by the PE configurations exhibit a marked increase, reaching levels approximately two orders of magnitude higher at 200 μsec. Below 15 μsec, the spectra for the PE and B_2O_3-PE configurations converge, with the PE configuration showing reduced intensity. The PE gamma-ray spectra appear dominant at around 1 μs in both figures. This was expected as prompt gamma rays travel at nearly the speed of light and thus arrive at the detector within a few nanoseconds (typically < 2 ns) over the 52 cm flight path. The sharp signal observed within 1 μs likely corresponds to these prompt gamma rays, which are produced from fast neutron interactions, particularly inelastic scattering, with hydrogen nuclei in the HDPE moderator. This interaction leads to a significant increase in gamma-ray flux within approximately 1 μs.

The gamma-ray contribution for B_2O_3-PE is higher than that of LiF-PE. This underscores the need to implement a detector capable of distinguishing between neutrons and gamma rays. Therefore, the experimental implementation focus will be on selecting a neutron detector with excellent neutron/gamma PSD capability in order to significantly mitigate the gamma-ray background by differentiating neutron signals from gamma-ray ones.

Both air (no material) and PE collimators are deemed unsuitable for qualitative and quantitative assessments utilizing the table-top NRTA system under development. A comparative analysis of the resonance dips (Fig. 2 a) from the simulations with both LiF-PE and B_2O_3-PE indicates that systems employing either collimator material are advantageous for qualitative NRTA evaluations. However, for quantitative assessments, a deeper resonance dip correlates with better resolution, and the B_2O_3-PE collimator exhibits superior performance in this regard. The forthcoming phase of this research project will concentrate on conducting preliminary experimental tests using the B_2O_3-PE collimator.

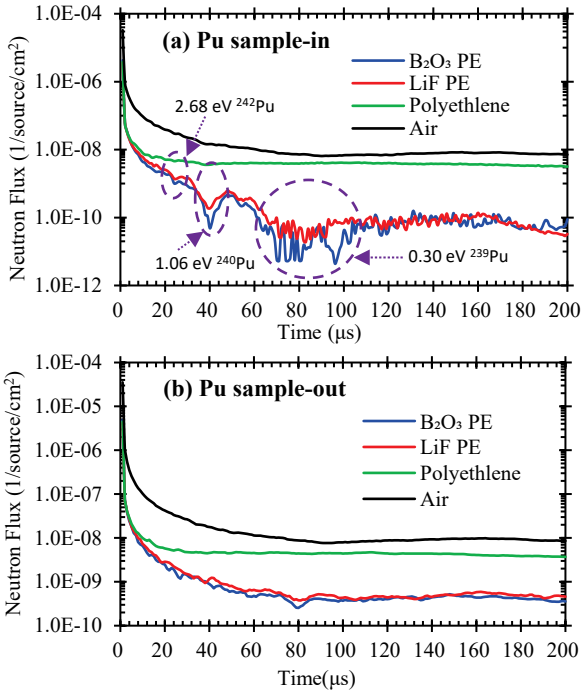

Fig. 2 Neutron TOF spectra for various collimator/shielding materials configurations under investigation: (a) sample-in spectra and (b) sample-out spectra.

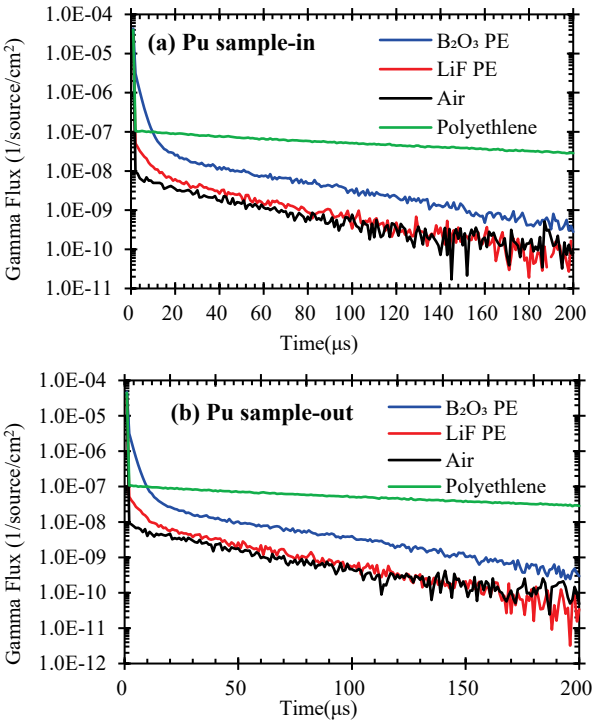

Fig. 3 Gamm-ray TOF spectra for various collimator/shielding materials configurations under investigation: (a) sample-in spectra and (b) sample-out spectra

V. Conclusions

The present study evaluates the performance of this new table-top NRTA system, which is currently under development. The development of the table-top NRTA system aimed at enhancing the practical utility of NRTA applications. The actual setup has a unique feature in its design as it does not rely on an accelerator or a deuterium-tritium neutron

generator tube as a neutron source. Instead, it utilizes a compact ^{252}Cf spontaneous neutron source. Additionally, it features a flight path length of less than 1 m (precisely 52 cm).

MC simulations were performed to calculate the neutron and gamma-ray fluxes produced by the system. The results indicated that the neutron fluxes exhibited resonance dips solely for two collimator materials - specifically, B_2O_3-PE and LiF-PE - when the Pu sample was present. In contrast, the neutron fluxes obtained with air and PE collimator materials were approximately an order of magnitude greater than those observed with the LiF-PE and B_2O_3-PE configurations, and resonance dips were absent in these cases. MC simulations have demonstrated the feasibility of NM detection with this tabletop NRTA system.

This study suggests that the tabletop NRTA system, employing the B_2O_3-PE as the collimator/shielding material is capable of detecting Pu nuclides in small samples of NMs, such as Pu pellets. Therefore, the experimental demonstration of the table-top NRTA system will use B_2O_3-PE as collimator material.

Acknowledgment

This work is supported by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT) under the subsidy for the "promotion of strengthening nuclear security and the like." This research was partly conducted with the supercomputer HPE SGI8600 in the Japan Atomic Energy Agency.

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