JAEA/ISCN Delayed Gamma-ray Spectroscopy Inverse Monte Carlo Development Status

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ABSTRACT

The Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) is developing Delayed Gamma-ray Spectroscopy (DGS) for safeguards capabilities. DGS is a non-destructive assay technique that utilizes neutrons to induce fission in the sample followed by a measurement of gamma rays emitted by the short-lived fission products as they decay. The primary DGS outcome is to evaluate the composition using the ratio of these gamma-ray peaks and the relative fissile-nuclide contribution. Significant progress has been made toward analyzing the gamma-ray peaks in order to evaluate the composition, as well as the mass, of fissile nuclides found in mixed nuclear material, like irradiated fuel. Specifically, the JAEA/ISCN is developing and inverse Monte Carlo (IMC) analysis method wherein the composite spectrum from a laboratory interrogation is compared to expected spectra from Monte Carlo (MC) simulations. Preliminary MC spectra show reasonable comparisons to measured spectra, though sufficient differences required further experimental confirmation. This work describes the recent efforts made to validate the JAEA/ISCN DGS MC through extended comparisons to measured spectra and other MC simulation programs. Further, we show how our MC comparison method has potential to assist in improving nuclear data useful for evaluating fission yields for nuclear safeguards and nuclear energy material accountancy.

Keywords: Delayed gamma rays, spectroscopy, Monte Carlo, simulation, high-radioactivity nuclear material

INTRODUCTION

Nuclear safeguards utilize verification of U and Pu materials to ensure the declared content exists [1-3]. Used nuclear fuel, a form of high-radioactive nuclear material, is particularly challenging due to the intense passive gamma-ray and neutron emissions from ^{137}Cs and minor actinides [4,5]. Present techniques for assemblies focus on gross- and partial-defect verification to ensure the item exists in the expected container and that the material shows signatures of being in a reactor [6-8]. Presently, partial defect verification utilizes correlations to ^{137}Cs and [134]Cs to determine relative burn-up, initial enrichment, and cooling time. Notably, Rokkasho Reprocessing Plant is expected to come online as the only reprocessing facility in a non-weapons state [9]. Consequently, safeguards were developed [10] to also include bias-defect verification of the input solution [8]. Specifically, Hybrid K-edge Densitometry and Isotope Dilution Mass Spectrometry (IDMS) are used to determine the U

and Pu masses and nuclide content, respectively. However, IDMS is a destructive analysis technique that additionally requires processing in the lab, uses limited reference materials, and takes a relatively long time to return a result [11].

To supplement used nuclear fuel safeguards, the Japan Atomic Energy Agency Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (JAEA/ISCN) is developing Delayed Gamma-ray Spectroscopy (DGS). DGS is an active-interrogation technique utilizing neutrons to induce fission followed by a measurement of gamma rays emitted as the resulting fission products decay. Importantly, the fission product contributions are specific to the fissionable nuclei undergoing fission and the relative contributions can be used to determine the nuclear material composition [12]. Applying DGS to used nuclear fuel, the low-energy passive gamma-ray emissions from ^{137}Cs must be suppressed to minimize detector dead-time. Consequently, we focus on the gamma rays with energy ≳2700 keV wherein the DGS signature is dominantly produced by short-lived fission products (e.g. ≲20 minutes). Subsequently, the signal strength must be increased, which is best performed with thermal neutrons, focusing the verification on fissile nuclides (see Fig. 1) [13].

The JAEA/ISCN DGS project began in 2015 [14] as a collaboration with the European Commission Joint Research Centre (EC/JRC). Over the past 8 years, multiple experiments have been performed with the Pulsed Neutron Interrogation Test Assembly (PUNITA) in the EC/JRC-Ispra (Italy) site [15]. Additional experiments were performed in the EC/JRC-Ispra PERLA facility, focusing on instrumentation development [16]. Separately, the JAEA/ISCN has been developing an inverse Monte Carlo (IMC) analysis method to evaluate the spectra. This work will describe the inverse Monte Carlo, summarize recent results, and highlight final development for this project.
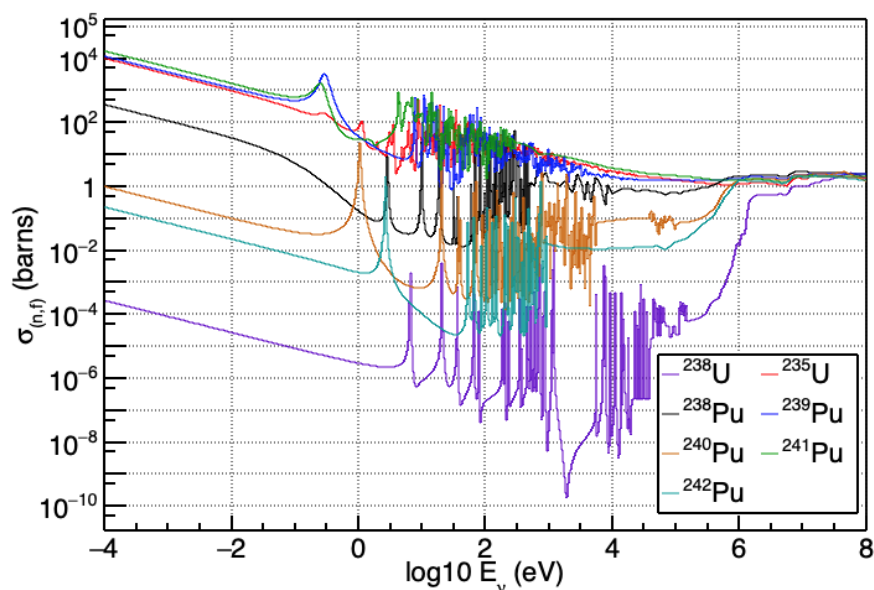

Figure 1. ENDF/B-VII.1 [13] neutron-induced fission cross-sections for the noted nuclides.

DGSMC INVERSE MONTE CARLO DEVELOPMENT

The DGS technique is ideally suited to verify the nuclear material composition by evaluating the fission product gamma-ray peak ratios that are directly correlated to the relative [235]U, [239]Pu, [241]Pu, and [238]U. It must be understood, though, that the DGS signature is affected by the method of interrogating the sample. For instance, neutron attenuation will occur in the container (e.g. reprocessing solution sample vial), as well as within the sample material. Additionally, gamma rays will be filtered not just by the gamma-ray detector shield, but also, again, within the sample and container. Finally, the spectrum will change over time from a combination both the half-lives of the fission products and the interrogation time pattern (irradiation, measurement, and cycles).

The JAEA/ISCN IMC analysis method compares spectra obtained from a real interrogation to a spectrum from simulations of the same interrogation derived from Monte Carlo. The simulations are created in the JAEA/ISCN Delayed Gamma-ray Spectroscopy Monte Carlo (DGSMC) using the ROOT analysis platform [17]. Users must input the interrogation pattern, sample conditions, instrument, detector, and shield of the real interrogation. From these inputs, the instrument defines the neutron spectrum derived from MCNP [18] and the detector defines both the efficiency derived from Geant4 [19] and the energy distribution from real detectors. The DGSMC code subsequently calculates the reaction rates, the number decays over each time period, and the gamma-ray observables from physics terms derived from nuclear databases [13,20]. The IMC analysis then compares the simulated peak ratios to the real peak ratios to determine the most-likely initial composition condition for verification purposes [21-23].

To improve the understanding of the expected spectra, we focused on two experiment types using PUNITA. First, experiments using only U or Pu samples allow us to understand fission product peak ratios directly with the dominant isotope, independent from other nuclide influences. Another experiment studied the how the [235]U and [239]Pu spectra changed due to the interrogation time pattern [24]. Specifically, due to the half-lives of the short-lived fission products, there will be an optimum interrogation pattern to be able to distinguish the [235]U peak ratios from the [239]Pu peak ratios. Nominally the optimum pattern should also distinguish the [241]Pu and [238]U contributions, though the samples available were not capable of providing this distinction. However, it was determined that shorter time patterns proved better, as highlighted in the change in the [95]Y 3575-keV and [91]Rb 3599-keV peak ratio and the number of gamma-ray peaks observed. This is a benefit since the shorter-lived nuclides are also more directly associated with the composition distinction from the independent fission yield and allow for quicker safeguards verification.

Notably, the same experiment resulted in showing a direct correlation between the [235]U mass and the integrated gamma-ray counts above 3300 keV [25]. This study improved upon earlier EC/JRC work [26] by showing ≲1.5% differences between quoted and evaluated masses. Specifically, this was due to improved fission rates and corrections to the neutron self-attenuation and gamma-ray self-shielding effects in the sample. Consequently, we were able

to show that fissile content (mass and composition) could be evaluated, with potential correlations to individual peaks using improved instrumentation.

The second type of experiments we performed were to study spectra related to the final application intent of determining the composition of mixed nuclear material samples. Since true mixed oxide samples were unavailable, our preliminary studies have focused on simulated mixed samples where we combined the mono-elemental U and Pu samples. During the first phase of development, we combined CBNM U samples of various enrichments [27] with PuGa samples of similar ^{239}Pu masses, focusing on maintaining the total fissile mass [28]. Spectral differences were observed, though precision measurements will be significantly improved with homogeneous samples, like in used fuel solution.

Recently we expanded the simulated mixed-sample study using the JAEA/ISCN Delayed Gamma-ray Test Spectrometer (DGTS) that uses ^{252}Cf as the neutron source [16]. We again combined the CBNM and PuGa samples, but instead focused on evaluating the effects of fixing either the ^{235}U or ^{239}Pu mass and increase the opposite. Though the ^{252}Cf intensity had decreased since the initial characterization of DGTS due to the COVID pandemic, we were still able to observe short-lived fission product gamma rays and differences from the changing masses. All of the measured spectra from this study, as well as the earlier studies, provide lessons on the optimum capability of a DGS interrogation.

JAEA/ISCN DGS MONTE CARLO VALIDATION

Using the previous experimental results, the JAEA/ISCN is currently in the process of evaluating the DGSMC for reliability within the IMC method. Specifically, we want to determine the quality of the simulated spectra, differences with short-lived fission product gamma-ray peaks, and calculation validation. Earlier studies indicated that there is a possibility of matching a measured spectrum to ~1% [21], though this was early and many spectra have been collected that can be used for validation studies.

We are presently finalizing a study of comparing the mono-nuclide spectra obtained for different interrogation patterns from PUNITA [24]. Specifically, we generated spectra using DGSMC for the same interrogation patterns and compare individual peak counts through trend and statistical tests to determine the relative quality (see Fig. 2). To validate the code, we also perform similar spectra to FIER [29] and FISPACT-II [30]. Individual Monte Carlo peaks vary significantly from the experiment for any given code, interrogation pattern, and sample, though improvements can be made with better experimental capabilities. However, it has been determined that the dominant difference is due to the fission yields consistently used across all codes.
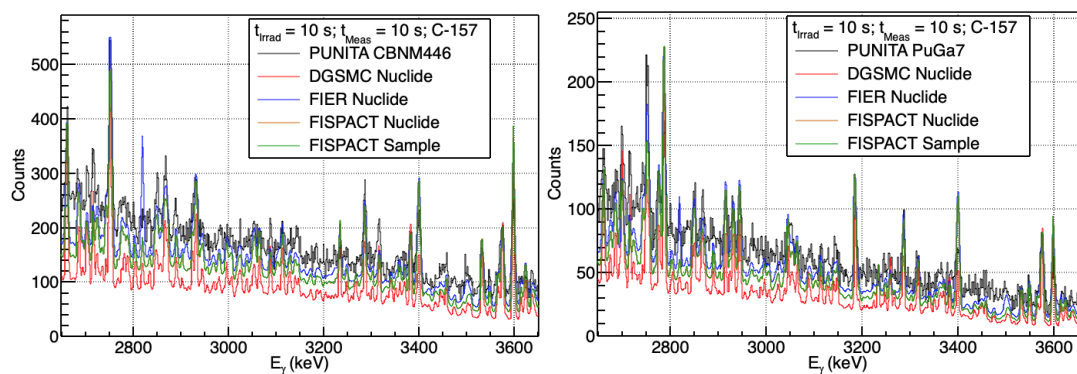
Figure 2. CBNM446 (^{235}U) and PuGa7 (^{239}Pu) measured spectra compared to simulations of the same interrogation listed in the legend.

FINAL ANALYTICAL DEVELOPMENT

The final development of the JAEA/ISCN goals are to integrate the analysis into the final instrument software and determine uncertainties on the evaluation for safeguards verification purposes [23]. This starts with improving the DGSMC code to calculate the Bateman equation directly, as is done in both FIER and FISPASCT [31]. Otherwise, DGSMC already includes calculation of activation products and has composition evaluation capabilities like FISPACT and calculates gamma rays emitted over the measurement period like FIER. Subsequently, studies can be made to improve the reliability of the simulated spectra for IMC analysis.

Due to the significant differences in the data-Monte Carlo comparison, the first studies will focus on variance reduction using multiple interrogations of the same sample. Beyond the reduction from multiple ^{235}U masses performed with the timing experiment [24], the JAEA/ISCN is upgrading the DGS laboratory with new U foils to perform this study (see Fig. 3). Once validating the method of improving the spectral reliability, experiments are under discussion with our EC/JRC collaborators to perform similar studies on the ^{239}Pu samples while seeking ^{241}Pu and mixed-oxide samples. These studies will be performed using the new JAEA/ISCN Fission Signature Assay Instrument (FSAI) (see Fig. 4) [32]. This new instrument was designed for reprocessing plant applications with small samples of used fuel solution, allowing the U foils to be evaluated well with the 3×10^8 n/s deuterium-deuterium neutron generator provided by Adelphi Technologies [33]. Notably, activation foils of similar dimensions to the U foils will be used to accurately characterize the neutron flux entering the nuclear material. Additionally, FSAI was designed to use multiple neutron detectors [34]: a ^{4}He detector [35] for source monitoring and two sets of ^{3}He detectors for prompt-neutron and delayed-neutron counting for supplemental mass evaluation.

The variance-reduction experiments will subsequently be used to evaluate the simulated spectra. DGSMC, FIER, and FISPACT will again be used, with a focus on improving the fission yield distributions by better aligning the simulated spectra to the highly accurate measurements. From these improved fission yields, the final optimization of the interrogation

pattern will be determined for highly-accurate safeguards verification results. Concurrently, multiple simulations will be run for each interrogation pattern to determine the smallest uncertainty in the composition and associated content evaluation.
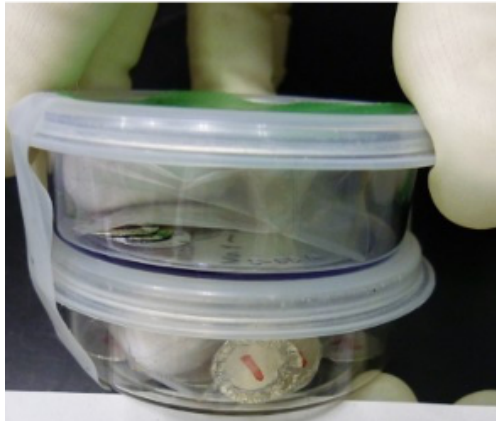

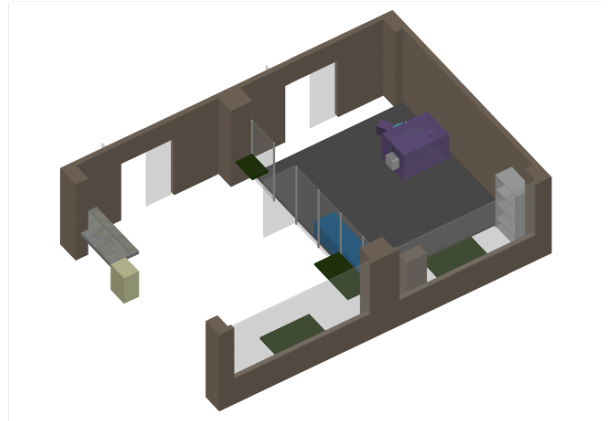Figure 3. JAEA/ISCN U foils to be used in spectral variance reduction studies.

Figure 4. JAEA/ISCN Fission Signature Assay Instrument in the DGS laboratory.

Final analytical development will focus on understanding conditions within final applications. For instance, past and future experiments focus on mono-elemental, solid samples, very different compared to reprocessing plants mixed-nuclide solutions. Assemblies have significant neutron self-attenuation and gamma-ray self-shielding due to the layering effects in the fuel rods, so we will expand upon our combined-sample studies to better understand these effects, including modeling these in MCNP. Finally, we will start investigations into Gen-IV fuels, like pebble-bed reactors and molten-salt reactors that are still under the safeguarding development stages [36,37] for which DGS may apply. Within this context, we will finalize the determination for IAEA verification goals concerning total Pu content and U enrichment [6,7,23].

SUMMARY

The JAEA /ISCN is developing an inverse Monte Carlo analysis method for the Delayed Gamma-ray Spectroscopy active-interrogation nondestructive assay technique. We utilize ROOT-based code to generate spectra from a simulation of actual interrogation conditions to determine the most-likely composition from spectral comparisons. Recent evaluations show data-Monte Carlo similarities are mostly dependent on peak intensities and resolution, but differences are largely due to fission yields in the nuclear databases. Improvements to the DGSMC code will focus on variance reduction and improved fission yield evaluations. Final development goals will then be to use DGSMC and other programs for final application possibilities and uncertainty evaluation..

security and the like". The authors would like to thank the EC/JRC collaborators for experiments highlighted in this work performed under the agreement between JAEA and EURATOM in the field of nuclear material safeguards research and development.

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