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JAEA-JRC Collaborative Development of Delayed Gamma-ray Spectroscopy for Nuclear Safeguards Nuclear Material Accountancy

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ABSTRACT

The Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) initiated a project to develop Delayed Gamma-ray Spectroscopy (DGS) in 2015. Concurrently, the Joint Research Centre of the European Commission (EC/JRC) joined the JAEA/ISCN in collaborative experimentation. The primary goal of the DGS collaboration is to enhance analytical capabilities and instrumentation to improve the material accountancy of mixed nuclear materials. Preliminary research focused on experimental evaluation to observe differences in standard U and Pu samples. Subsequent efforts focused on understanding the correlated instrumentation to obtain viable signatures to distinguish the fissile nuclide composition. Our current efforts are to finalize the instrumentation and associated analysis required to efficiently determine the mass content in irradiated fuel. This work will describe the efforts made in this JAEA/ISCN-EC/JRC collaboration to enable DGS to become a viable non-destructive assay method for safeguards capabilities. Specifically, we will describe the lessons learned used for the final JAEA/ISCN instrument design from experiments performed in EC/JRC Ispra, analytical verification and model design validation experiments performed jointly in EC/JRC Geel, and spectral evaluations performed in both EC/JRC and JAEA/ISCN laboratories.

Keywords: Safeguards, NDA, delayed gamma rays, spectroscopy, fission, high-radioactivity nuclear material, used nuclear fuel

INTRODUCTION

Safeguards are enabled to ensure the peaceful use of commercial nuclear materials [1-3]. Verification within safeguards agreements depend on the precision level required to reach conclusions that the U and Pu exists as declared by the state and operator [4,5]. High-radioactivity nuclear material (HRNM) like used nuclear fuel is primarily verified for gross defects, for instance counting items and checking for damage of the vessel.

Additionally, partial defect verification is performed by looking at general quantity differences of U and Pu in materials, usually using a nondestructive assay (NDA) method [6].

Used nuclear fuel assemblies have different U and Pu contents that change over time relative to the initial enrichment, burn up, and cooling. Due to the inability to observe the U and Pu directly [7], inspectors usually perform correlation calculations with relative ^{137}Cs and ^{134}Cs gamma rays in the ~600-keV energy range. Contrarily, in reprocessing plants where fuel material is removed from the assemblies the U and Pu is separated, more stringent methods are required to verify the nuclear material [4]. The input dissolution usually undergoes Hybrid K-Edge Densitometry and Isotope Dilution Mass Spectrometry (IDMS) [5,8]. These bias defect verification measures allow inspectors to observe small differences in the declarations to ensure quantities are not diverted over long periods of time from this more accessible form. However, IDMS is a destructive analysis method requiring laboratory separation and it takes a long time to return a result on a limited number of samples [9]. Additionally, fuels from newer reactor designs are still undergoing safeguards development designs and new verification techniques may be required [10,11].

Addressing some of the limitations, the Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) has been investigating active-interrogation NDA techniques [12]. The JAEA concurrently established collaborative research into this active-NDA program with the European Commission Joint Research Centre (EC/JRC). This research is intended to supplement or introduce new methods to evaluate HRNM by inducing fission in the fissile U and Pu nuclides. Of these techniques, the Delayed Gamma-ray Spectroscopy (DGS) technique has the potential to evaluate the fissile nuclide ratios for determining the fissile composition [13,14]. Specifically, the DGS technique utilizes an interrogation sequence of irradiating a sample to create fission products, followed by a measurement of the gamma rays emitted by the fission products following well-defined decay chains.

In this work, we describe the project goals and activities of the JAEA-JRC DGS collaboration. First, we will highlight the past developments from Phases I and II in which we focused on basic studies of fission product gamma rays and instrumentation development. We will then discuss the past, current, and future efforts of this current Phase-III, focusing on recent joint experiments.

PAST DEVELOPMENT

The DGS project started in 2015 with Phase-I focusing on basic experiments to observe fission product spectra from U and Pu [15]. Within these studies, multiple ^{235}U and ^{239}Pu masses were measured to look for differences in relative peak intensities. Notably, preliminary studies investigated combining different mass samples in order to evaluate composition distinction (see Fig. 1a). These experiments were performed using the EC/JRC-Ispra (Italy) PUNITA instrument that was designed for differential die-away analysis using a deuterium-tritium (D-T) neutron generator [16]. These experiments were subsequently used to start the validation of the JAEA/ISCN DGS Monte Carlo (DGSMC) [17] being developed in the ROOT analysis platform [18].

Within the perspective of instrumentation development, PUNITA was adapted to enable a standard DGS interrogation (see Fig. 1b). The final configuration consisted of one lower ^{3}He detector bank panel replaced by a high-density polyethylene (HDPE) panel with a hole to allow the sample to be transferred to the detector below the instrument. This allowed the detector to remain mostly outside of the neutron field, shielded by the remaining moderator materials, while maximizing the thermal neutron flux closer to the sample in the cavity.

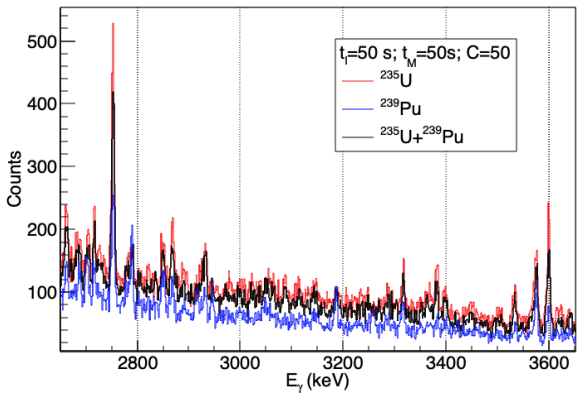

Figure 1a. Delayed gamma-ray spectra from individual and combined Pu and U samples interrogated with a 50-s irradiation and measurement pattern over 50 cycles.

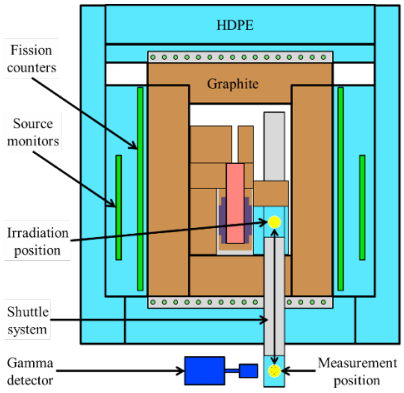

Figure 1b. PUNITA adapted for a DGS in interrogation with the cavity filled with graphite and the sample transferring the sample to the detector below.

In Phase-II, lessons learned from Phase-I enabled significant progress for both instrumentation and analysis development. First, the JAEA/ISCN Delayed Gamma-ray Test Spectrometer (DGTS) was characterized and tested for DGS capabilities using ^{252}Cf sources in EC/JRC-Ispra PERLA facility [19]. DGTS was designed similar to PUNITA with a graphite core, but was initially designed to transfer the source. Progress showed sample-transfer, like that of PUNITA, is better for radioisotope sources due to the hazards and environmental activation of the source being outside of the moderator. Subsequent instrumentation further investigated moderators with only HDPE for direct comparisons to DGTS [20,21].

Phase-II experiments focused on improving the analysis capability and effects from the sample. Utilizing PUNITA in the final Phase-I configuration, U and Pu samples were interrogated with multiple neutron irradiation and gamma-ray measurement times [22]. These time differences showed significant differences in which fission product gamma rays appeared due to the various half-lives of the short-lived fission products (see Fig. 2a). Comparing the small set of U and Pu spectra, we determined a preliminary optimum interrogation time pattern to distinguish ^{235}U from ^{239}Pu of 60-s for both irradiation and measurement times over a 1-hour interrogation. Concurrently, the same U samples were used to evaluate mass correlations from increasing ^{235}U enrichments [23]. Corrections to the neutron self-attenuation, gamma-ray self-shielding, and D-T temperature variation resulting in correlations with the mass to ~1% error. Notably, these correlations were independent of

the interrogation time pattern of the previous study (see Fig. 2b), indicating that not just composition, but also fissile nuclide content could be determined from a DGS interrogation.

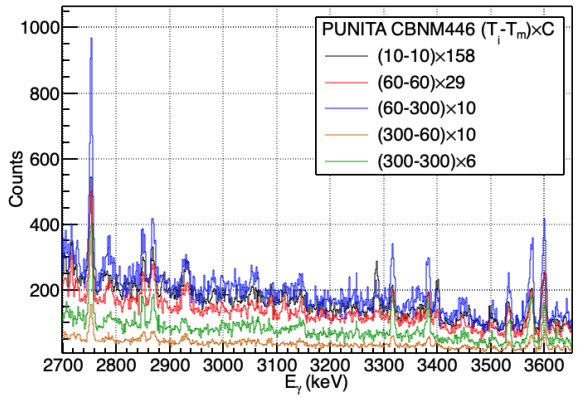

Figure 2a. Delayed gamma-ray spectra from U samples interrogated with multiple interrogation time patterns as labeled.

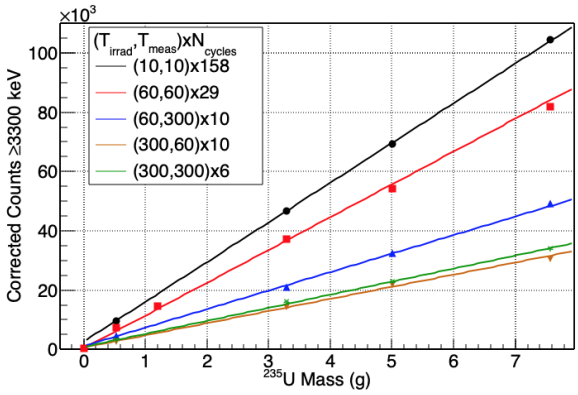

Figure 2b. Linear correlations between sample ^{235}U masses and delayed gamma-ray counts for the time patterns in Fig. 2a.

PHASE-III RECENT ACTIVITIES

Presently, the DGS project is in the third and final phase of development in which we are focusing on final integration between the analysis and instrument. Specifically, the JAEA/ISCN is developing the Fission Signature Assay Instrument (FSAI) that incorporates all of the lessons learned from previous phases for an optimum instrument to evaluate small samples using a deuterium-deuterium (D-D) neutron generator. Additionally, activities are underway to finalize the development of the JAEA/ISCN DGSMC for inverse Monte Carlo analysis.

Within the instrumentation development, two experiments were performed recently in the EC/JRC-Geel (Belgium) MONNET tandem accelerator facility. First, the ^{4}He detector intended for FSAI source monitoring was characterized for energy dependence [24]. Final efficiency characterization is currently underway for proper source scaling as was performed with PUNITA. Next, an experiment was performed to validate the MCNP [25] models created toward the final design of FSAI [26]. The JAEA/ISCN Test Moderator consisted of variable HDPE and graphite pieces to evaluate the moderator effects on the neutron flux evaluated through count-rate differences in ^{3}He detectors (see Fig. 3). Various challenges in the experiment required additional moderating materials to be used, expanding the capability for model comparisons for both D-T and D-D neutrons. Final analysis is underway, focusing on beam-current intensity correlations.

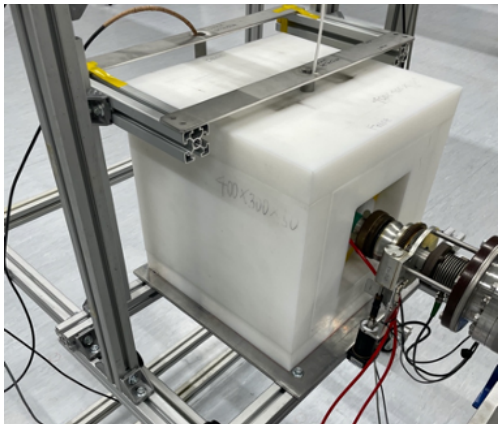

Figure 3. JAEA/ISCN Test Moderator in the experimental position in front of the MONNET beam head.

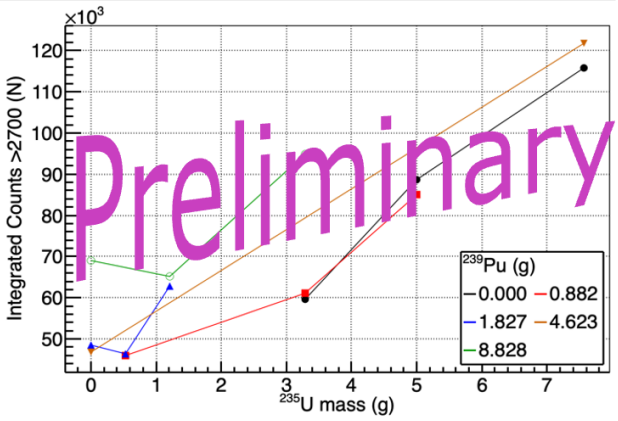

Figure 4. Preliminary correlation between ^{235}U mass gamma-ray counts for fixed ^{239}Pu samples interrogated with DGTS in PERLA.

Toward analysis development, an experiment was performed with DGTS in PERLA expanding upon the Phase-I mixed sample studies. Specifically, we focused on fixing the ^{235}U enrichment of the CBNM samples [27] or the ^{239}Pu PuGa samples and evaluating the mass correlations to the opposite sample (see Fig. 4). Final analysis is underway, including evaluating the capacity to observe gamma-ray peak ratios to determine composition differences of the sample combinations.

PHASE-III FINAL DEVELOPMENT

As mentioned, the final Phase-III goals will focus on the integration between instrumentation and analysis. Primarily this must focus on the optimization of the interrogation to distinguish not just ^{235}U from ^{239}Pu, but also ^{241}Pu expected in used nuclear fuel. Toward this goal, JAEA/ISCN will characterize the FSAI neutron flux in the sample space, the ^{4}He source monitoring, and ^{3}He detectors incorporated for supplemental prompt and delayed neutron signatures [28]. Concurrently, EC/JRC-Ispra PERLA is also installing a D-D neutron generator instrument to expand their capabilities, include those for the DGS project [29]. The PERLA D-D instrument will incorporate the full-HDPE design from previous studies and utilize a horizontal track to shift the sample from inside of the cavity to the side of irradiator, similar to early PUNITA studies [30].

Both of these instruments can be used to finalize the DGS analysis goals. From the JAEA/ISCN side, we are preparing for potential applications to reprocessing plant input accountancy dilutions. Previous studies highlighted the differences between combined samples and expected homogenous samples. However, significant differences between the observed spectrum and DGSMC simulated spectrum indicate individual spectra must be better understood from both enrichment and interrogation pattern perspectives [31]. Subsequently, plans are being finalized for using FSAI and the EC/JRC D-D instrument to study variance reduction and improved simulations. In particular, JAEA/ISCN has obtained new U foil samples to study the variations on enrichment, attenuations, and sample effects; discussions are underway to perform similar studies on Pu samples in PERLA.

Additional JAEA/ISCN goals are to apply DGS toward used nuclear fuel assemblies and subsequent investigation is underway on practical instrumentation [32]. Previous combined-sample studies exemplify the effects of self-attenuation and self-shielding potentially observable from assemblies, and investigations are underway to evaluate instrumentation constraints. Toward this goal, discussions are underway regarding the potential to apply the DGS technique to ^{241}Pu samples to improve the analytic evaluation from used nuclear fuel assemblies and solutions.

Finally, studies will be performed to evaluate neutron signatures concurrently obtained during a DGS interrogation. In particular, the ^{3}He detectors incorporated in the FSAI irradiator will enable prompt fission neutrons to be observed during the irradiation period. Additional ^{3}He detectors located near the gamma-ray detector could concurrently observe possible delayed neutrons emitted by some short-lived fission products for improved mass evaluation. Experiments are also under discussion to perform similar interrogations of U and Pu samples with PUNITA in order to validate and expand the FSAI results.

SUMMARY

The JAEA /ISCN and the EC/JRC are collaborating in the development of delayed gamma-ray spectroscopy to supplement present safeguards verification. Significant progress was made over the previous two phases toward both instrumentation and analytical development. This third phase focuses on optimizing the analysis and evaluating final instrumentation, as seen in recent experiments for both instrumentation and mixed nuclear material analysis development. Final goals will focus on integrating the analysis capability into the instrument constraints for optimal interrogation and verification capabilities.

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