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Development of Delayed Gamma-ray Spectroscopy for Nuclear Safeguards (1): Project Overview

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ABSTRACT

Present safeguards verification of spent nuclear fuel solution in reprocessing plants utilizes mainly destructive analysis techniques to obtain the nuclide content. However, alternative non-destructive assay (NDA) methods could help to improve the number of verified samples and the speed of verification and reduce generated waste and reference material dependence. Toward this goal, the Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) started developing Delayed Gamma-ray Spectroscopy (DGS) as an NDA technique. DGS 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. Our parallel primary goals are to efficiently analyze the sample and to develop a compact instrument to be easily installable into analytical laboratories. Starting in 2015, the JAEA/ISCN initiated a collaboration with the European Commission Joint Research Centre (EC/JRC) to collect DGS experimental data. The JAEA-JRC collaboration began testing JAEA-developed DGS instruments in 2018 to determine requirements of the final design for deployment. This work will describe the status and overall progress of this DGS development followed by a discussion of future plans toward final deployment. Highlights of each development phase will be provided as introductions to the other presentations in this series, including a description of our next demonstration workshop.

Keywords: delayed gamma rays, nuclear safeguards, active neutron interrogation.

INTRODUCTION

Nuclear power is of significant importance for many nation states to maintain viable energy capabilities. In order to prevent the diversion of nuclear material to develop weapons, the International Atomic Energy Agency (IAEA) employs safeguards agreements with those states [1,2,3]. Within those agreements, the state must account for all of the nuclear material in their facilities. The IAEA would then verify this declared amount through defect analysis on the various items, including taking samples, and deploying containment and surveillance measures. Primarily, the IAEA must quantify the amount of U and Pu [4], usually through measurements of neutrons and gamma rays emitted during the nuclide's decay.

However, this is particularly challenging for spent nuclear fuel found throughout much of the nuclear fuel cycle. Specifically $10^{5\text{-}8}$ neutrons/second and $10^{13\text{-}17}$ gammas/second are passively emitted from minor actinides and fission products, depending on burn-up, initial enrichment, and cool-down [5]. Between this and the fact that the fuel assembly is effectively sealed, most spent nuclear fuel is verified as an item, only checking for radioactivity. The U and Pu is determined by the operator using burn-up code calculations but not explicitly measured [4,6].

Reprocessing plants are an exception to this method of verification since the U and Pu is separated from the single item form factor. Quantitative verification only starts after the separation process begins, once the assembly is cut apart and the spent nuclear fuel is dissolved [7]. High-precision measurements of the spent nuclear fuel dissolution are made since the U and Pu can be directly measured on samples taken from holding tanks. Specifically, Hybrid K-Edge Densitometry (HKED) [8,4] is utilized to determine the absolute elemental masses and Isotope Dilution Mass Spectrometry (IDMS) for nuclide content [4,7]. The nuclear material masses are then taken as absolute and back-reported for shipper-receiver difference calculations. It should be noted though, that IDMS is a destructive analysis method requiring pre-processing of the sample that extends the time to return a report and generates waste in the laboratory [9]. Further, it requires reference materials for calibration that are becoming limited in availability.

Much research has gone into improving the verification of spent nuclear fuel [10]. One promising technique is delayed gamma-ray spectroscopy (DGS), an active interrogation technique that can be used to evaluate the primary fissionable nuclides (U-235, Pu-239, Pu-241, and U-238). Specifically, DGS uses the gamma rays emitted by short-lived fission products generated during neutron-induced fission to evaluate the relative quantities of the fissionable nuclides. This is comparable to using the long-lived fission products (e.g. Cs-137), but is an improvement since the short-lived fission products are more distinctive to the fissionable nuclide (see Figure 1). The technique first uses an external neutron source to induce fission in the sample. After the irradiation, the generated fission products decay proportionally to their half-lives, the fission yield, and the relative amounts of the fissionable nuclide.

To perform the irradiation and measurement interrogation, different instrumentation is required depending on the sample type and space available. Early research focused on applying DGS to full assemblies, primarily in storage ponds [11]. However, due to the lower passive emission rates, it is easier to apply DGS to reprocessing plant input solutions since smaller amount of detector shielding are required [12,13]. Further, it is possible to apply this toward reprocessing plant in-line verification [14], either at the input solution tank or along the pipes. Regardless of the application, though, the analysis must be independent of the sample type and instrumentation.

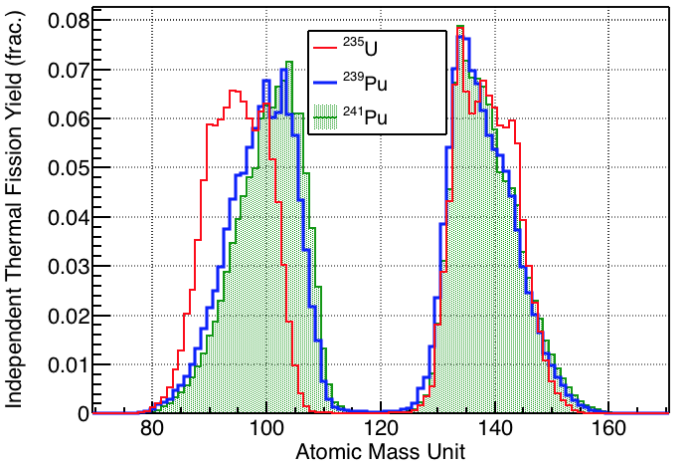

Figure 1. Mean thermal fission yields of ^{235}U, ^{239}Pu, and ^{241}Pu from the ENDF/B-VII.1 database.

JAEA-JRC DGS PROJECT

The JAEA/ISCN initiated a program to investigate promising NDA techniques for NM detection and analysis [15,16]. This program includes multiple neutron and gamma signature techniques, but DGS became a stand-alone project due to the relative technical readiness level. The JAEA/ISCN focused on developing a general analysis development and designed experiments to observe differences in U/Pu spectra. To help perform the basic research to improve the capability, this program was started as a collaboration with the European Commission's Joint Research Centre (EC/JRC). Within the collaboration the EC/JRC has hosted the JAEA/ISCN for the experiments since nuclear material standard samples are readily available. EC/JRC has multiple neutron sources and have extensive experience performing neutron irradiation experiments.

The collaboration is mostly driven by the JAEA/ISCN MEXT subsidy, with the JAEA/ISCN goal of demonstrating the DGS capability as a viable option for safeguards verification. Within this context, the first-stage development goal is to show that DGS can be performed on small samples of mixed nuclear material, like spent nuclear fuel solution in a reprocessing plant. Toward this goal, the JAEA/ISCN is performing parallel development of the analysis and instrumentation. The primary scope is to improve the capability to quantify the fissionable nuclide content within spent nuclear fuel. Dominantly this focuses on the relative content (composition) of the U and Pu nuclides for which gamma rays are useful. Additionally, since the amount of gamma-ray signal is dependent on the amount of nuclear material, there is also a mass correlation perspective that is being investigated. The other JAEA/ISCN DGS project scope is to develop a DGS instrument that can be used on the small samples and that will add additional constraints to the analysis. From this preliminary foundation, the JAEA/ISCN is investigating the application of DGS to other forms of spent and mixed nuclear fuel [11,17].

PREVIOUS RESULTS

The JAEA/ISCN-EC/JRC DGS project was planned for three multiple year-long phases. Phase-I occurred between the 2015 and 2018 Japanese fiscal years (April 1-March 31). The primary scope was to perform basic DGS measurements of fissionable materials. Measurements were performed with PUNITA [18] at the Ispra, Italy site of EC/JRC. Standard U [19] and Pu samples were interrogated to test the possibility to observe fission product gamma rays compared to earlier research [20]. The observation of these delayed gamma rays encouraged the JAEA/ISCN to perform additional experiments with gradual improvements. In parallel the JAEA/ISCN started to

develop an analysis capability using an inverse Monte Carlo method. This was designed to perform a standard DGS interrogation utilizing various nuclear data inputs to calculate the reaction rates through the gamma-ray detection. Initial results showed good reliability [21], but further experiments were required to validate the code. Phase-I ended with a demonstration of the experimental results, Monte Carlo description, and outlook for designing a compact DGS instrument.

Phase-II started in April 2018 and will end in March, 2022. Primary work has focused on improving a compact DGS instrument tested in PERLA of EC/JRC-Ispra. This Delayed Gamma-ray Test Spectrometer (DGTS) uses Cf-252 inside HDPE and graphite moderator to irradiate the sample and a linear robot to transfer the sample to the detector. The initial design of DGTS focused on not moving the sample due to facility restrictions. This required the source to be shuttled while trying to optimize the neutron flux and gamma-ray detection without damaging the HPGe detector. Since the delayed gamma-ray signals were not observed, the JAEA/ISCN redesigned DGTS to move the sample, improving the fission rates and gamma-ray collection. As part of these efforts, the EC/JRC evaluated a pure HDPE moderator flux was evaluated using activation foils with the JAEA/ISCN supporting the MCNP model comparison [22,23]. Additional work has been underway in the JAEA/ISCN to quantify the effects that various moderator materials have on the flux in compact conditions using Cf-252 sources.

Beyond developing DGS instrumentation, additional experiments have been performed to improve the analytic capabilities. Returning to PUNITA, the U and Pu samples were tested under tightly constrained conditions to study the timing sensitivity of the DG spectra. These allowed the JAEA/ISCN to expand the capability to distinguish U and Pu compositions [24] and also to perform mass correlations [25,26] (see Figure 2).

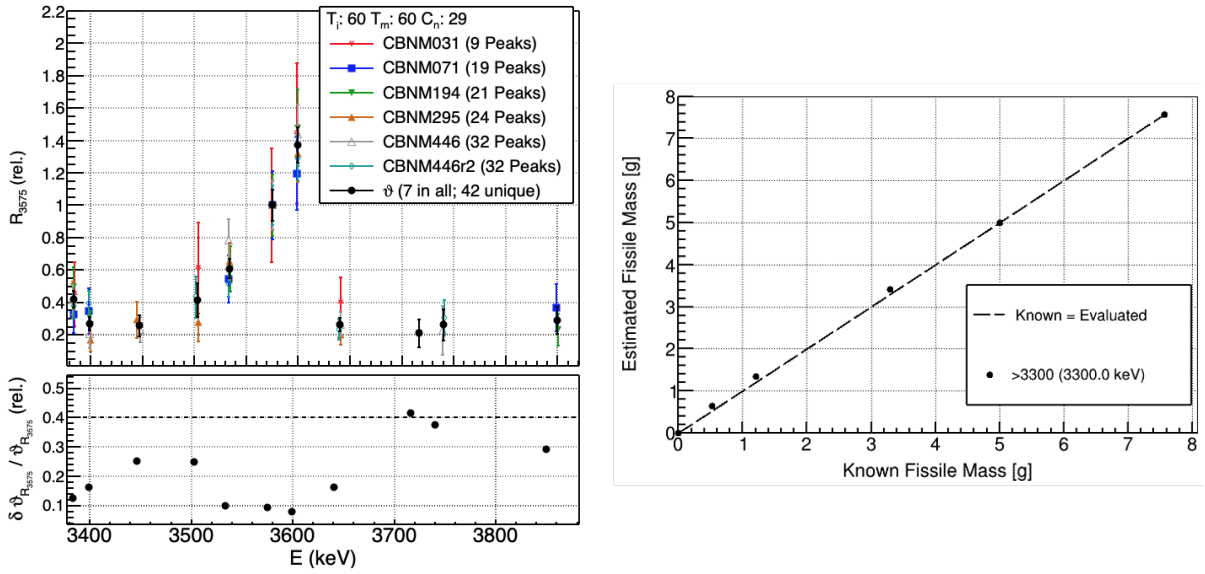

Figure 2. Fission product gamma-ray peak ratio for composition analysis (left) and integrated counts for mass correlation analysis (right).

FUTURE PLANS

In this last year of our Phase-II development we will hold another demonstration of our progress and future direction. The remaining development goals focus on improving the analysis using the

JAEA/ISCN Monte Carlo (DGSMC) and testing instrument sensitivities on the observed signatures. DGSMC improvements will be made through comparison to the recent PUNITA timing measurements. Simulations will then be performed to evaluate expected signatures of mixed nuclear materials.

To improve the instrumentation, the JAEA/ISCN is introducing neutron monitoring that will be tested in a new irradiator at PERLA after characterizing it in the JAEA. Concurrently, additional studies will be performed in preparation of Phase-III. Specifically, experiments will be performed with the MONNET tandem accelerator of the Geel, Belgium site of the EC/JRC. These will focus on evaluating the differences between Cf-252 and D-D and D-T neutron sources for final DGS instrumentation. The results will be used for model validation and an optimized DGS instrument design. Finally, MONNET will be used to evaluate JAEA/ISCN neutron detector response and characterization that will be incorporated as neutron monitoring and signature evaluation.

The third phase of this DGS project will start in April, 2022 and focus on final analysis and instrument development for application studies with mixed nuclear material. First, the JAEA/ISCN will incorporate all of the lessons learned from Phase-I and Phase-II into an optimized DGS instrument for small sample verification (see Figure 3). This will focus on

1) sample, neutron source, and moderator effects on the fission rate;
2) sample, detector, and filtering effects on the gamma-ray measurement; and
3) sample transfer, timing, and neutron monitoring effects on the analysis.

Complimentary requirements will be made for the use of this in a facility, focusing on dose minimization, contamination control, and maintenance. We will characterize the instrument, quantifying sensitivities of the neutron generator, gamma-ray detector, and the sample. Once characterized, working with the EC/JRC, the instrument is expected to be deployed to various facilities to evaluate representative samples for calibration. Additionally, final JAEA/ISCN quantification analysis will proceed based on the final instrument capabilities. This will include simulating the expected gamma-ray spectrum based on instrument timing and flux constraints.

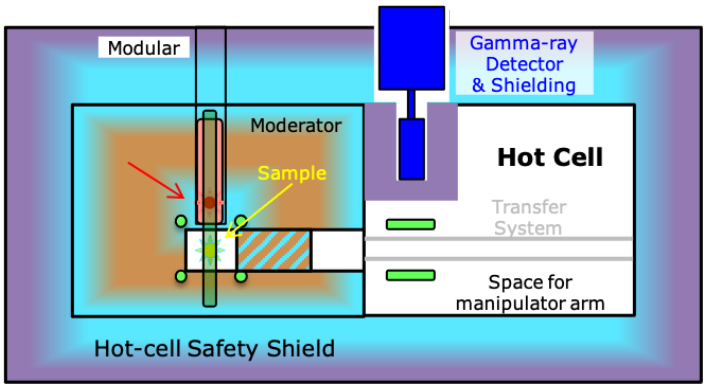

Figure 3. Preliminary concept for the JAEA/ISCN practical DGS instrument.

Beyond this, evaluations will be made on how to apply this JAEA/ISCN-EC/JRC capability. For instance, though this is already underway, the JAEA/ISCN will finalize the evaluation for spent nuclear fuel assemblies. This will be evaluated for those found in reactors and reprocessing plants, but also those found in short- and long-term storage. Further, we will investigate applying this to alternative fuel cycles (e.g. ADS, MSR) and in-live verification.

Present studies show that DGS is largely limited by the fission yields in nuclear data, with which the JAEA/ISCN DGSMC can potentially help improve [27]. For these improvements, discussions are currently underway with the EC/JRC on possible experimental locations and materials. This would then enable the improvement of burn-up codes, understanding heat variation in reactors, and even the source terms for reactor neutrinos dependent on the beta decay of these generated fission products [28].

SUMMARY

The JAEA/ISCN and EC/JRC are collaborating on the development of DGS for nuclear safeguards capabilities. Over the past year, advances have been made within this project to improve the analytic quantification of the composition and mass of fissile nuclides. Additionally, progress has been made toward refining the instrumentation required to perform the interrogation of small mixed nuclear material samples. Future efforts will focus on optimizing the analytic and instrument capabilities and evaluate applications beyond reprocessing plant input solution samples.

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