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Delayed Gamma-ray Assay for Nuclear Materials Safeguards. Comprehensive Technology Readiness Assessment

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Delayed Gamma-ray Assay for Nuclear Materials Safeguards

Comprehensive Technology Readiness Assessment

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Summary

Spectroscopic analysis of beta-delayed gamma-rays emitted from fission products following active interrogation was investigated as a non-destructive assay technique for detection and characterization of nuclear materials in shielded and complex configurations. This measurement technique is expected to be applicable to a variety of applications within the nuclear material safeguards, arms control, and emergency response areas, where active neutron interrogation is acceptable.

Potential applications of the delayed gamma-ray assay in the nuclear material safeguards area include characterization of U and Pu isotopic content in spent and unirradiated nuclear fuel, enrichment measurements, verification of bulk material in containers, and the characterization of nuclear wastes. In the arms control area, this method can be used as a part of warhead dismantlement measurements to confirm separation of components, and may be applicable in warhead confirmation. In the context of emergency response, it can be used for detailed diagnostics of an unknown item.

At present, the delayed gamma-ray assay technology reliably qualifies for the Technical Readiness Level 3 (TRL 3). The R&D work completed under this project has successfully demonstrated the empirical proof-of-concept for this active interrogation technology for a variety of measurement configurations, neutron sources, and nuclear material samples. A robust first-principles analytical signature modeling methodology was developed, experimentally validated, and can be easily extrapolated to a range of realistic measurement scenarios. Several response analysis methodologies have been implemented and deemed reliable for a variety of active interrogation applications in the areas of nuclear material safeguards, arms control, and emergency response. The initial readiness level before the start of this project was assessed at TRL 0, since only fragmentary records of the signature observations and speculative statements of the relevant non-destructive assay applications existed at that time.

The delayed gamma-ray technology readiness can be easily advanced to TRL 4 in a short period, if a certain measurement application is specified and set for a demonstration in a laboratory environment.

The critical hardware components of the delayed gamma-ray measurement technology (neutron generators, gamma-ray spectrometers) are readily available as COTS and can be configured for a specific deployment configuration.

The primary risk of the delayed gamma-ray assay methodology is defined by the application-specific uncertainties. Evaluation of the system performance can be effectively completed once

the expected measurement purpose, nuclear material characteristics, dimensions, and radiation exposure constraints are specified. A minor risk can be associated with the delayed gamma-ray response analysis techniques. Several approaches were proposed and demonstrated primarily using the modeled signatures. The observed performance may not be easily transferrable to the conditions of a real measurement setup, and some additional R&D may be required.

Overall, the delayed gamma-ray assay technology can be recommended for follow-up demonstrations addressing relevant measurement applications in the areas of nuclear material safeguards, arms control, and emergency response.

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1. Project Overview

1.1 Project Objectives

Spectroscopic analysis of beta-delayed gamma-rays emitted from fission products following active interrogation was investigated as a non-destructive assay technique for detection and characterization of nuclear materials in shielded and complex configurations. This measurement technique is expected to be applicable to a variety of applications within the nuclear material safeguards, arms control, and emergency response areas, where active neutron interrogation is acceptable. Delayed gamma-ray measurement can be deployed as part of an integrated active interrogation system that would evaluate a range of complementary neutron induced signatures, such as prompt gamma-ray and neutron analysis, delayed neutron, neutron die-away, neutron transmission analysis, and even neutron and gamma-ray imaging. Delayed gamma-ray assay evaluates direct signatures specific to isotopes that undergo induced fissions under active interrogation. It can be used to determine the relative isotopic abundance in a mixed nuclear material, detect the presence of a specific fissionable isotope in the presence of other fissile materials, or, in a calibrated setup, determine absolute isotopic amounts.

Potential applications of the delayed gamma-ray assay in the nuclear material safeguards area include characterization of U and Pu isotopic content in spent and unirradiated nuclear fuel, enrichment measurements, verification of bulk material in containers, and the characterization of nuclear wastes. In the arms control area, this method can be used as a part of warhead dismantlement measurements to confirm separation of components, and may be applicable in warhead confirmation. In the context of emergency response, it can be used for detailed diagnostics of an unknown item.

The delayed gamma-ray assay technique is based on neutron induced fission that generates a gamma-ray response from short-lived fission fragments with half-lives ranging from seconds to tens of minutes providing “fingerprints” of the fissionable isotopes. Each fissionable isotope produces characteristic relative peak intensities so that the measured delayed gamma-ray spectrum can be analyzed as a superposition of contributions from individual isotopes. Isotopic fractions can be determined if the response spectra of the individual isotopes are accurately known.

1.2 Project Description

Investigation of the delayed gamma-ray signatures included two key components: a strong experimental campaign to characterize the delayed gamma-ray signatures of the isotopes of interests and of combined targets, and a closely linked modeling effort to assess system designs and applications. Experimental measurements were performed to evaluate fission fragment yields, to test methods for determining isotopic fractions, and to benchmark the modeling code package. Detailed signature knowledge is essential for analyzing the capabilities of the delayed gamma technique, optimizing measurement parameters, and specifying neutron source and gamma-ray detection system requirements.

Experimental measurements of the delayed gamma-ray responses were carried out at the Idaho Accelerator Center (IAC) using an accelerator-driven photo-neutron source and at the Lawrence Livermore National Laboratory (LLNL) using portable DT and DD neutron generators. At the IAC, a high-intensity neutron interrogation setup was optimal for the characterization of high-energy delayed gamma rays from small ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu targets. Data was collected for pure and combined targets for several irradiation/spectroscopy cycle times ranging from 10/10 seconds to 15/30 minutes. A plethora of delayed gamma ray lines from short-lived fission products was identified in the 3-6 MeV energy range and examined for potential use in delayed gamma assays. The ^{235}U/^{239}Pu line ratios vary considerably with several lines much stronger for ^{235}U fission. For shorter irradiation/spectroscopy periods, two lines of ^{106}Tc with a half-life of 36 s are prominent in the ^{239}Pu spectra but nearly absent in the ^{235}U spectra and thus strongly indicate the occurrence of ^{239}Pu fission. Experimental measurements at LLNL demonstrated that the delayed gamma-ray response can be reliably obtained from shielded nuclear material samples with a mass of 100 grams and above using a DT or DD neutron source with an output of 1e8 neutrons per second and above.

An extensive delayed gamma-ray response modeling methodology was developed in the course of the project, and was used to simulate passive, prompt, and delayed gamma-ray spectra replicating the experimentally collected data. This modeling capability was utilized to analyze measurement results and assess application-specific assay concepts including the assay simulation of complex objects and regimes (including multiple irradiation/spectroscopy cycles). The code package was benchmarked against the data collected at the IAC for small targets and the data from larger (> 100 gram) samples of nuclear materials collected at LLNL. The modeling results indicate that delayed gamma-ray responses can be reliably collected from a variety of nuclear material objects with statistical quality sufficient for analyzing their isotopic composition using COTS neutron generators and detector instrumentation.

Delayed gamma-ray response de-convolution analysis was developed and demonstrated for experimentally observed and modeled spectra. A number of delayed gamma-ray peaks in the energy region up to 4.5 MeV were identified as sensitive to the fissionable isotopic content of the assayed nuclear material, and modeled responses were used to demonstrate determination of the relative fissile isotopic compositions. When applied to the analysis of simulated spent nuclear fuel assemblies, this method demonstrated that assembly-averaged relative fissile isotopic content can be determined with uncertainties on the order of 5 to 10% for ^{239}Pu and ^{235}U. A major contribution to the uncertainty can be attributed to the fact that the detectors are mostly sensitive to the outer few rows of pins while the burnup and thus the isotopic concentrations may vary considerably across an assembly. Although the methodology for the absolute normalization of the response has not been fully developed, this analysis technique demonstrated strong potential, and may be used in combination with other measurements to achieve highly accurate fissionable material composition characterization.

The primary conclusions from this experimental and modeling effort can be summarized as follows:

- Delayed gamma-ray assay following an active interrogation with a neutron source is a feasible method for nuclear material safeguards and is applicable to quantitative analysis complex and shielded objects such as a spent nuclear assembly.
- Delayed gamma-rays emitted from nuclear materials can be detected with adequate accuracy during minutes following the active interrogation and are directly proportional to the abundancy of fissionable isotopes in the investigated object.
- Response de-convolution methods that determine the relative composition of fissionable isotopes in the interrogated material have been demonstrated in the course of the project. This methodology can be utilized as a part of the integrated assay approach to determine the absolute isotopic and elemental composition of spent fuel assemblies and bulk material.
- A comprehensive modeling methodology for simulating delayed, prompt, and passive gamma-ray responses in active neutron interrogation was developed in the course of the project. This methodology was experimentally validated with accessible nuclear material samples and used to extrapolate analysis to complex systems such as spent nuclear assemblies.
- During experimental validation of the modeling methodology, significant disagreements for isolated delayed gamma-ray lines were observed. Discrepancies were attributed to values in the ENDF/B-VII.1 nuclear data library with a recommendation to reduce the uncertainties in energy-dependent fission yields, gamma-ray yields, and branching ratios in the future.
- Delayed gamma-ray assay requires an interrogating neutron source with high output, at least 10^{11} n/s intensity is required for characterization of spent nuclear fuel assemblies. For unirradiated nuclear materials, a portable neutron source with the output of 10^{8} n/s and above is required. These source intensity requirements should be revised and adjusted for the specific measurement scenarios and configuration.

- The measurement methodology is expandable to other applications, such as non-destructive assay of bulk nuclear material, arms control and emergency response applications.

The original project findings and recommendations were related to the Next Generation Safeguards Initiative that considered a range of candidate passive and active interrogation techniques for spent fuel characterization prior to disposition in a deep underground repository.

As a direct consequence of the original project, the delayed gamma-ray assay is currently considered as one of the methods for the analysis of the reactor debris in the Fukushima accident remediation effort. This methodology is actively pursued by the Japanese Atomic Energy Agency (JAEA) as a part of the integrated debris evaluation station that also involves Neutron Resonance Transmission Analysis (NRTA), Differential Die-Away (DDA) analysis, and Prompt Gamma-ray Analysis (PGA).

The modeling methodology that was developed for predicting the delayed gamma-ray responses was adapted for the design and performance evaluation effort for the Passive Gamma-ray Emission Tomography (PGET) instrument investigated by the International Atomic Energy Agency (IAEA) for the partial/bias defect detection in spent fuel assemblies. Major elements of the modeling technique were transferred to the IAEA as a part of the PGET Performance Prediction (PPP) toolkit developed collaboratively by Lawrence Livermore and Pacific Northwest National Laboratories in 2018.

1.3 System Description

The delayed gamma-ray measurement technique utilizes an external neutron source (D-T or D-D generator, or a small accelerator-driven photo-neutron source), and gamma-ray spectrometers to perform characterization of SNM materials behind shielding and in complex configurations such as a nuclear fuel assembly. Delayed gamma-ray spectra are recorded with a high-resolution spectrometer following an active neutron interrogation period. Analysis of peak ratios and intensities in the recorded spectra provide a signature for detecting the presence or absence of nuclear materials in shielded enclosures, identifying specific isotopes in the mixture, and determining the relative composition of fissionable isotopes in the mixture.

Potential safeguards applications of this method are: 1) characterization of fresh and spent nuclear fuel assemblies in wet or dry storage; 2) analysis of uranium enrichment in shielded or non-characterized containers or in the presence of a strong radioactive background and plutonium contamination; 3) characterization of waste and product streams at nuclear material processing plants. Other applications can include warhead confirmation and warhead dismantlement confirmation in the arms control area, as well as SNM diagnostics for emergency response needs.

In its current state, the delayed gamma-ray measurement method qualifies for TRL-3 level with a proven physics concept, demonstrated measurement technique, response sensitivity and analysis approaches. Empirical and modeling studies were completed in support of this SNM characterization technique. Extensive experimental tests involving weapons-grade Pu, HEU and depleted uranium samples were completed. Elements of the specifically developed modeling code were benchmarked during passive high-resolution gamma-ray measurements of spent fuel at the CLAB facility in Sweden.

Compared to traditional nuclear material assay methods (passive gamma-ray spectroscopy, total and time-correlated neutron counting) the delayed gamma-ray technique is less sensitive to shielding, the presence of an intense gamma-ray background, and provides SNM isotope-specific signatures. The delayed gamma-ray measurement technique is well suited for deployment as part of an integrated active neutron interrogation system that can exploit complementary signatures form prompt gamma-ray, delayed and die-away neutrons, neutron resonance transmission analysis, active source imaging, etc.

2. Project Technology Risks Summary and Readiness Assessment

2.1 Critical Technology Elements

Experimental measurements completed during the original delayed gamma-ray assay project were carried out primarily at the Idaho Accelerator Center using a photo-neutron source to characterize the emission of high-energy, beta-delayed gamma-rays from ^{235}U, ^{238}U, ^{239}Pu, and ^{241}Pu targets following neutron induced fission. Data sets were collected for pure and combined nuclear material targets for several irradiation/spectroscopy cycle times ranging from 10/10 seconds to 15/30 minutes. This experimental setup is well suited for the fundamental analysis of active interrogation signatures, as it provides a wide range of controls over the timing, energy, and intensity parameters of the interrogating neutron source. However, because of the facilities and infrastructure required for the linear accelerator that drives the neutron production, this setup is hardly transferrable to a field application.

An important observation from the earlier effort documented discrepancies for some of the characteristic gamma-ray lines in the delayed gamma-ray spectra obtained experimentally and produced by the modeling technique. These discrepancies were attributed to neutron fission yield data, as well as gamma-ray yield and branching ratios for some of the fission products as recorded in ENDF/B-VII.1, ENSDF, and auxiliary nuclear data libraries.

The primary configuration constraints for the delayed gamma-ray assay in the context of nuclear material safeguards, arms control, and emergency response applications are summarized below.

> *Purpose of the measurement.* The delayed gamma-ray setup complexity changes considerably depending on the desired functionality. In its most complex form, it should support determination of relative content of the fashioning isotopes in the measured object. This will require the highest intensity of the neutron source coupled with a high-resolution gamma-ray spectrometer, and a fixed setup. The opposite simple task of determining the presence of a fissionable material behind shielding, may be accomplished with a smaller source and a medium-resolution spectrometer (such as $LaBr_3$). Other measurement objectives may include confirming uranium enrichment, determining material amount, detecting a certain fissionable isotope in the presence of a known one, etc. Specifying the purpose for the active interrogation measurement is a crucial design and performance evaluation component.

> *Nuclear material composition.* The expected nature of the nuclear material to be measured is another critical constraint. Analysis of irradiated material will require additional shielding of the spectrometer and will limit the response analysis to a higher-

energy region, potentially increasing the neutron source intensity requirement. More complex material mixtures (for example a combination Pu with U with a range of enrichments) will require a more elaborate setup.

Nuclear material amount, shielding, and packaging. Naturally, this is an important application-specific constraint that will affect the measurement setup configuration. Evaluating larger items (approximately 100 grams or more) will offer more flexibility with the placing and intensity of the neutron source, and the choice of the gamma-ray spectrometer. Shielding affects both the propagation of the interrogating neutrons through the item, and their moderation. Smaller effective packaging offers options for a more compact measurement setup.

Target assay time. As demonstrated in the original project, the gamma-ray response structure is affected by the interrogation time pattern. Depending on the target assay time, the measurement may be designed for the "one-pass" assay (single irradiation period followed by a single detection period), or for a "pulsed" one (with seconds- or minutes-long interrogation/detection periods).

Neutron source placement and intensity limit. Position of the neutron source relative to the interrogated item is a major factor. A mere standoff distance is a direct constraint on the intensity of the neutron source. Ability to incorporate neutron moderating and reflecting materials around the neutron source may be beneficial in certain applications, but is not always available in the field. ALARA radiation exposure considerations are also critical, as is the placement of the measurement setup. The source intensity must also account for exposure to nearby personnel.

Gamma-ray spectrometer(s) placement. The intensity and resolution of the delayed gamma-ray spectra is a critical component of the response analysis and the primary focus of the application-specific measurement setup design. While it is desirable to ensure the highest possible detector efficiency to the nuclear material, count rate constraints require elaborate shielding and collimation. It is crucial to minimize neutron transport from the source to the spectrometer during the interrogation in order to avoid damage and activation. If the source intensity during interrogation is high, negative effects were observed as a result of neutron scattering from the interrogating object towards the detector. Such a situation may require a shutter or a motion setup to protect the spectrometer.

The above measurement setup constraints are recursively related and complex. As a result, the practicality of the delayed gamma-ray assay should be investigated in the application-specific format, when the expected functionality and items are specified.

2.2 Technology Risk Assessment

In order to expand the delay gamma-ray assay methodology to specific applications within the nuclear safeguards, arms control and emergency response area, it is important to determine whether it can be implemented in the field setup using COTS neutron sources and detectors. A series of experiments completed so far prove the overall feasibility of this non-destructive assay technique, however its exact performance depends on the particular measurement scenario.

The primary risk with the delayed gamma-ray assay methodology can be associated with application-specific uncertainties, such as the required nuclear material sensitivity threshold, the expected amount, composition, and packaging of the material, and the maximum permissible neutron source output rates (radiation exposure rates in the measurement area). Since the signature collection relies on COTS components, existing neutron generators and high-resolution gamma-ray spectroscopy hardware is sufficient to perform the measurement. The specific application will dictate how effectively this hardware can be packaged around the inspected objects, and will determine the desired operational parameters (neutron output, interrogation time pattern, detector efficiency and count rate limits).

In the course of conducting the experimental delayed gamma-ray measurements, the primary complexity that the research team encountered was optimizing the placement and shielding of the gamma-ray spectrometer used in the delayed gamma-ray assay. This element of the interrogation setup design is not easily modeled and optimization between the geometric efficiency to the interrogated object and the detector shielding and collimation was most successful when completed empirically. Neutrons from the interrogating source or scattered from the measured object may affect the operation of the spectrometer. Excessive activation of the nuclear material is also detrimental for delayed gamma-ray spectroscopy as it increases dead time and causes pile-up effects. The highest quality gamma-ray spectra produced in this project were obtained when nuclear material samples were manually transferred between the irradiation and spectroscopy setups.

A minor risk with the delayed gamma-ray assay is present in the response analysis technique. The resulting spectroscopic signature incorporates contributions from all fissionable isotopes present in the interrogated item. Therefore, a response de-convolution analysis is required to determine the exact relative abundances and absolute amounts of contributing U and Pu isotopes. Several of these methodologies have been investigated in the course of the project, some relying on the calibration of the empirical signature, and some arising from the first principles. These analysis techniques were tested against a limited set of experimental data, and were primarily demonstrated using extensive series of modeled spectra. It is possible that the successful performance observed in the simulated environment may not be easily transferrable to the conditions of a real application-specific measurement setup. Moreover, the nuclear library data (fission product yields, branching ratios, etc.) used in a subset of the candidate de-convolution approaches may not adequately reflect the entire reality of the active interrogation or may have considerable associated uncertainties.

2.3 Technology Demonstration in a Relevant Environment

Based on the series of experimental and modeling investigations completed so far, the delayed gamma-ray assay technique can be strongly recommended for a follow-up demonstration in a relevant, application-specific environment. The critical hardware components (neutron sources, gamma-ray spectrometers) are readily available in COTS configuration, and fully adequate for the response collection. Modeling methodology has been demonstrated to accurately predict signatures, and can be effectively used to assess measurement configuration, instrument design, and scenario-specific response analysis development.

The next series of the delayed gamma-ray assay technique development should be performed for a specific, well defined application. Once the measurement objectives and constraints are clearly identified, they will provide a set of target parameters for which the measurement setup will be defined. The delayed gamma-ray interrogation concept has been demonstrated to be flexible and adaptable to various measurement configurations, sources, and nuclear material characteristics. This project has provided sufficient tools to support a low-cost evaluation of the optimal setup requirement for each application-specific measurement requirement.

The technology demonstration scenarios, that can be recommended for a near-term implementation, should include applications that can be tested in a laboratory, and then be promptly transferred to a relevant environment. Therefore, immediately pursuing the interrogation of spent nuclear fuel assemblies may not be optimal, since the entire complexity of the realistic measurement parameters and constraints cannot be easily replicated outside of the actual spent fuel handling facility.

In the context of nuclear material safeguards, the delayed gamma-ray assay technique can be readily demonstrated for the uranium enrichment measurement behind unknown shielding or in a complex configuration, including the UF_6 storage cylinders. It can also be readily applied to the characterization of bulk material containers (for example, MOX powder, LEU powder or pellets, unirradiated fuel assemblies). It can also be deployed for the characterization of waste packages to determine the TRU content and composition. Additionally, the technique can be used to detect the presence of a fissionable isotope even if its characteristic signature is obstructed by another expected material in a shielding or masking configuration.

For arms control applications, the delayed gamma-ray assay may be demonstrated for the warhead dismantlement confirmation, where it can be used to characterize components containers emerging from the disassembly process. It is an effective technique for determining the presence or absence of nuclear material in shielded containers, where it can achieve considerably lower minimal detectable mass amounts than conventional passive techniques.

In the emergency response area, the delayed gamma-ray technique can be demonstrated for the diagnostics of a suspect object enclosed in a container with an unknown configuration (a drum). This is especially relevant for current studies in neutron transmission imaging that consider the deployment of an intense neutron imaging source in the field.

3. Conclusions

At present, the delayed gamma-ray assay technology reliably qualifies for the Technical Readiness Level 3 (TRL 3). The R&D work completed under this project has successfully demonstrated the empirical proof-of-concept for this active interrogation technology for a variety of measurement configurations, neutron sources, and nuclear material samples. A robust first-principles analytical signature modeling methodology was developed, experimentally validated, and can be easily extrapolated to a range of realistic measurement scenarios. Several response analysis methodologies have been implemented and deemed reliable for a variety of active interrogation applications in the areas of nuclear material safeguards, arms control, and emergency response. The initial readiness level before the start of this project was assessed at TRL 0, since only fragmentary records of the signature observations and speculative statements of the relevant non-destructive assay applications existed at that time.

The delayed gamma-ray technology readiness can be easily advanced to TRL 4 in a short period, if a certain measurement application is specified and set for a demonstration in a laboratory environment.

Other practical outcomes from this project include recommendations to the Next Generation Safeguards Initiative effort on active interrogation of spent fuel assemblies, input to the non-destructive assay methods evaluation for the Fukushima debris analysis, and the expansion of the modeling technique to enable gamma-ray source and transport calculations for the Passive Gamma-ray Emission Tomography (PGET) project.

The critical hardware components of the delayed gamma-ray measurement technology (neutron generators, gamma-ray spectrometers) are readily available as COTS and can be configured for a specific deployment configuration.

The primary risk of the delayed gamma-ray assay methodology is defined by the application-specific uncertainties. Evaluation of the system performance can be effectively completed once the expected measurement purpose, nuclear material characteristics, dimensions, and radiation exposure constraints are specified. A minor risk can be associated with the delayed gamma-ray response analysis techniques. Several approaches were proposed and demonstrated primarily using the modeled signatures. The observed performance may not be easily transferrable to the conditions of a real measurement setup, and some additional R&D may be required.

Overall, the delayed gamma-ray assay technology can be recommended for follow-up demonstrations addressing relevant measurement applications in the areas of nuclear material safeguards, arms control, and emergency response.

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