Proceedings of the INMM & ESARDA Joint Virtual Annual Meeting
August 23-26 & August 30-September 1, 2021

Development of Delayed Gamma-ray Spectroscopy for Nuclear Safeguards (4): Integrated Neutron Detection Systems

Hee-Jae Lee
ISCN, Japan Atomic Energy Agency
Tokai-mura, Ibaraki, Japan

Mitsuo Koizumi
ISCN, Japan Atomic Energy Agency
Tokai-mura, Ibaraki, Japan

Douglas Chase Rodriguez
ISCN, Japan Atomic Energy Agency
Tokai-mura, Ibaraki, Japan

Fabiana Rossi
ISCN, Japan Atomic Energy Agency
Tokai-mura, Ibaraki, Japan

Tohn Takahashi
ISCN, Japan Atomic Energy Agency
Tokai-mura, Ibaraki, Japan

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. Our earlier experimental results show that delayed gamma-ray spectra can be used to estimate fissile mass using neutron source normalization. As such, the JAEA/ISCN is developing a compact DGS instrument that will incorporate a neutron detection system of multiple neutron monitors to improve the accuracy of mass evaluation. Specifically, the JAEA/ISCN is investigating ^{3}He and ^{4}He neutron detectors for both thermal and fast neutron evaluations. Constrained by the compactness requirement, the detectors' response for short source distances have been studied. This paper will introduce the scope of neutron detection system followed by a description of the detector characteristics and response at short source distances, including that related to our demonstration irradiator. Finally, preliminary integrated neutron detection systems within a compact DGS instrument will be discussed.

Introduction

As required by the nonproliferation treaty [1], the IAEA introduces safeguards to verify that fissile materials (U and Pu) are not diverted for nuclear weapon production. In the reprocessing plant, spent nuclear fuels are converted into a solution form that can have a higher risk of diversion. To safeguard against this, the IAEA monitors the solution throughout the facility, including taking samples to quantify the U and Pu [2]. In an effort to improve sample verification, the Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (ISCN) of the Japan Atomic Energy Agency (JAEA) is developing the Delayed Gamma-ray Spectroscopy (DGS) technique [3]. DGS is a non-destructive assay method that utilizes neutrons to induce fission and generate fission products in the nuclear material sample. These fission products decay and emit a unique gamma-ray spectrum that can be used to determine the isotopic composition of the sample. Due to the high-intensity passive emission from spent nuclear fuel, DGS focuses on the high-energy beta-delayed gamma-rays from short-lived fission products.

The JAEA/ISCN DGS instrument is being designed for compactness around a neutron irradiation system and a gamma-ray detection system. The neutron irradiation system consists of a fast-neutron source and a moderator to reduce the energy for high thermal fission rates. After irradiation, the gamma-ray detection system measures the sample, possibly after transferring it to a different location. This interrogation pattern is generally repeated to reach a minimum statistical level.

The final JAEA/ISCN instrument will use a neutron generator because it is easier to handle and does not provide activation background during the measurement periods [4]. However, neutron emission rates can vary with the operating temperature as observed in PUNITA experiments [5]. This became important for mass correlation where the number of gamma-rays depends upon the fissile mass and source intensity. If it is found that ^{252}Cf should be used, though it generates neutrons continuously, the absolute emission rates would be required for proper scaling. By introducing neutron detection systems, DGS instrument will be able to confirm source consistency during irradiation. Moreover, the neutron detection systems can be used as the neutron signature of fission occurring in the sample. First, the prompt fission neutrons can be detected above ~2.5 MeV or will provide excess counts above the ^{252}Cf source. Second, delayed neutrons can be collected concurrently with delayed gamma rays because they are come from decay of some fission products, providing redundant fission signatures. Using these two neutron signatures for mass evaluation in conjunction with the delayed gamma-ray mass evaluation will improve the accuracy by reducing the variance.

The JAEA/ISCN is investigating ^{4}He and ^{3}He neutron detectors for our final DGS instrument. The ^{4}He detector is a fast neutron detector, intended to be used as the source monitor because the signal has a correlation with the incident neutron energy. Regarding a Deuterium-Deuterium (D-D) neutron generator, this energy discrimination has the advantage of separating prompt fission neutrons from the source neutrons. The signal from prompt fission neutrons for mass evaluation will be collected using the ^{3}He detectors due to their high efficiency. The ^{3}He detectors will also be used for the delayed neutron detection system. This paper will describe the characterization studies of those detectors and then how they will be introduced as neutron detection systems in our DGS instrument.

Detector Characterization

The purpose of this section is to describe the characterization of the ^{4}He and ^{3}He neutron detectors in relation to a compact DGS instrument.

^{4}He Neutron Detector

The JAEA/ISCN purchased a ^{4}He detector for the DGS instrument since it is the least sensitive to gamma rays of all fast neutron detector types. This is important because there is a significant prompt gamma-ray background during the irradiation that would be observed concurrently. An Arktis Radiation Detectors Model S670 [6] was chosen considering the length of the detector (~90-cm long) and the compactness of the DGS instrument. In the tube, the ~200 atm ^{4}He gas interacts with incident neutrons through elastic scattering [7], which results in scintillation light generation. This scintillation light is detected with 12 silicon photomultipliers (SiPM) on printed circuit board strips that send the signals to a readout board assembly. A unique characteristic of this detector is that the SiPMs are optically separated into three segments that can be used independently.

The ^{4}He data acquisition system required high sampling rates to separate signal pulses due to the high flux expected from the external neutron source and the induced fission neutrons. As such, an ADQ14 data acquisition unit from Teledyne SP devices [8] converts the S670 analog signals into digital pulses at a 1GHz sampling rate. This provides a 0.125-ps time resolution for the beginning time stamp of each waveform (see Figure 1a), which is usually collected with 14-bit, 1-ns time bins and a 16-bit pulse amplitude range. The acquired digital waveform is then processed by python code with true signals minimally determined using a >200 ns Time-over-Threshold (ToT) discrimination cut (see Figure 1b).

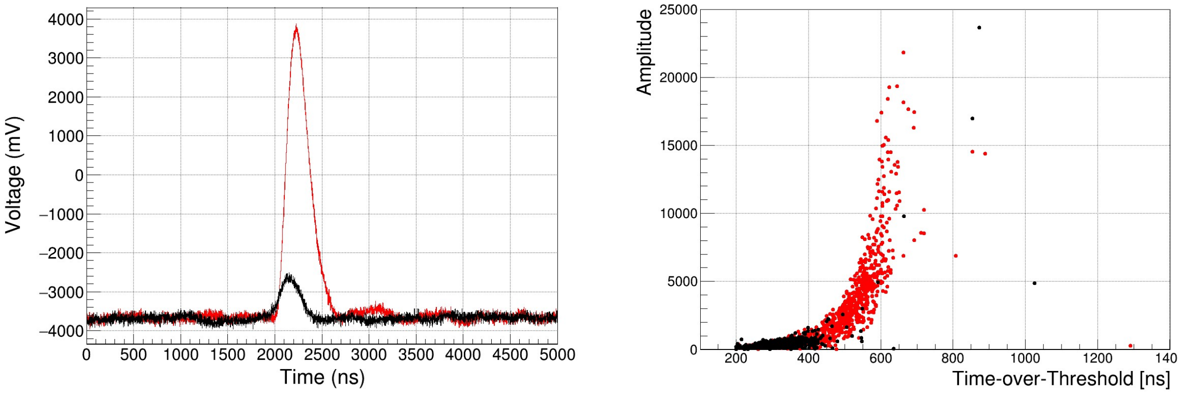
To distinguish neutron signals from gamma ray signals, the waveforms were evaluated for pulse shape such as rise-time and ToT using standard radioisotope sources (e.g. ^{252}Cf, ^{137}Cs, and ^{60}Co). The ^{4}He elastic cross-section is expected to show energy dependence with the incident neutron [9], therefore, we are investigating how to use the pulse amplitude for calibration. The detection profile was measured by moving a ^{252}Cf source along the detector length and counting the number of interactions in each segment (see Figure 2). This measurement shows segment separation and that the neutron signals are dominant in the segment in front of the source. These measurements were performed with the source in contact with the detector and more investigation is needed.

^{3}He Neutron Detector

Again considering compactness in a final DGS instrument, the JAEA/ISCN purchased small ^{3}He detectors from General Electric (GE) Reuter Stokes [10]. These contain 10 atm of ^{3}He gas inside a tube that is 18 cm long and 1.27 cm in diameter. The signal is read out using an Easy-MCS [11], limiting the count rates to ~10^{5} cps that must be considered in the high thermal flux present during the DGS interrogation.

The ^{3}He detectors were characterized in the thermal neutron field of a high-activity ^{252}Cf source inside a graphite pile at the Facility of Radiation Standards (FRS) [12]. They were placed at a distance of 120 cm from the source to determine the detector efficiency. The detector sensitivity profile used in the efficiency calibration was evaluated using collimation with Cd tubes around the ^{3}He detector at the same position [13]. The Particle and Heavy Ion Transport code System (PHITS) code was used to validate the sensitive region and input rates for the efficiency by comparing the results with the measurements.

Noting that the detector will be used in a compact DGS instrument, the ^{3}He detector was re-tested with a 3-cm source-detector distance and a low-activity ^{252}Cf sources. Similar to the FRS profile test, the ^{3}He detector was scanned along a 1-mm slit collimator behind different thickness of High-Density Polyethylene plates in the space between source and detector. Using the validated ^{3}He detector model, PHITS simulations were performed using both ^{3}He(n,p) and ^{3}He(n,tot) cross-sections with the ^{3}He(n,tot) in better agreement to the measurements. This indicates that elastic scattering becomes important since many fast neutrons contributed to the signal through down-scattering within the detector to reach the (n,p)-dominant energy region. These high-pressure detectors must be evaluated for positions within the compact instrument since they will experience high fast-neutron fluxes and retain more neutrons.



(a) **(b)**

Figure 1. Digitized waveform (a) and pulse-height vs. TOT distribution (b) of neutrons (in red) and gamma-rays (in black).

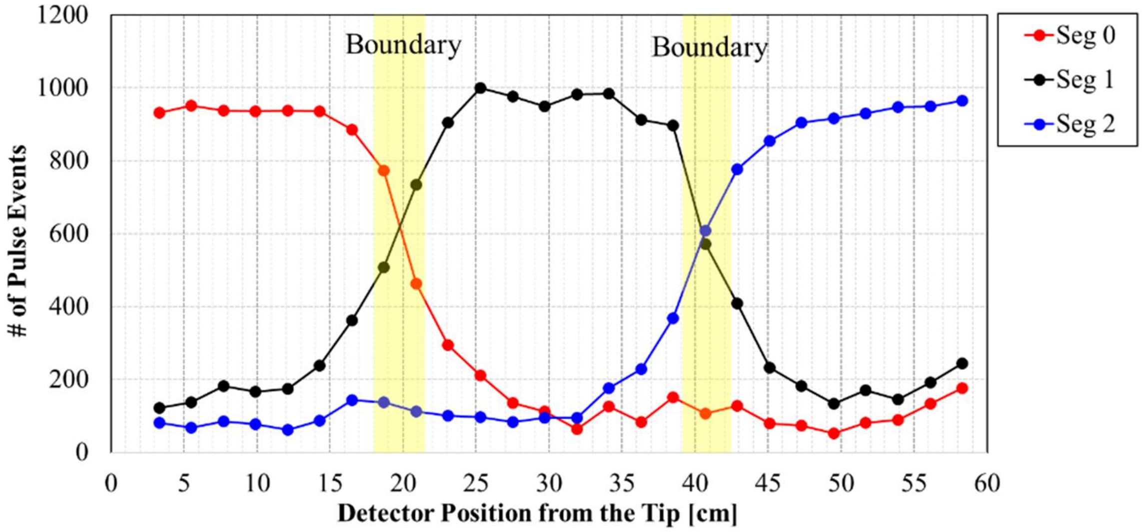


Figure 2. Preliminary study of the number of neutron signals acquired for each detector position.

Integrating Neutron Detection into Final Instrument

Studies are currently underway to optimize the incorporation of these detectors into the compact DGS instrument (see Figure 3a). For the source monitoring system, an end segment of the ^{4}He detector will be placed over the neutron source emission position, for instance ^{252}Cf capsule or neutron generator target. During the irradiation, the sample will also be positioned in this segment, measuring sufficient rates from both source and sample. Consequently, the other end segment will be used as the source monitor since the signal will be dominated by source neutrons. In the case of a D-D generator, the source neutrons are emitted only during the irradiation period and at a maximum 2.45 MeV energy with down-scattering in the moderator. Monitoring this mono-energetic peak will provide correlation to the emission rate. The source monitor can be used to determine the ^{252}Cf emission rate during the measurement period when the sample is not contributing to the signal. Though ^{252}Cf emits neutrons continuously, monitoring it allows the inspector to ensure the correct fission rate scaler is applied for the expected activity.

In order to monitor the D-D emission peak, energy and efficiency calibration of the ^{4}He detector must be performed. The JAEA/ISCN performed initial tests using the AmBe source of the FRS and is planning to use the FRS Deuterium-Tritium accelerator configuration. Additional calibration studies will carried out in collaboration with European Commission Joint Research Centre in Geel (Belgium) using the MONNET tandem accelerator. In parallel to the energy calibration, the JAEA/ISCN is optimizing the detector positioning in the final instrument using MCNP models.

For prompt fission detection, a ^{3}He neutron detector system will be located near the sample irradiation position. During the irradiation period, prompt fission neutrons are the excess counts from the source neutron background from either a D-D generator or ^{252}Cf. Notably, this excess will also appear in the ^{4}He detector with the additional benefit that we can discriminate the prompt fission neutrons as those above ~2.5 MeV from a D-D generator. A separate ^{3}He detector system will be placed near the measurement position to collect delayed neutrons concurrently with the gamma rays as the fission products decay. With a D-D generator, there will be no source neutrons emitted during the measurement period. However, there may be continuous background with ^{252}Cf that must be removed.

In our development progress, the JAEA/ISCN manufactured a new irradiator for our end-of-phase workshop that includes ^{3}He detectors to monitor the source and sample neutrons. Presently, the JAEA/ISCN is characterizing this new irradiator for detector responses and neutron flux including how the detectors interfere with each other and the sample space. Within this irradiator, we will perform additional detector studies using ^{3}He detectors with different pressure or volumes compared to the presented above. Further studies will include our sensitivity to differentiate between source neutron and fission neutron signatures.

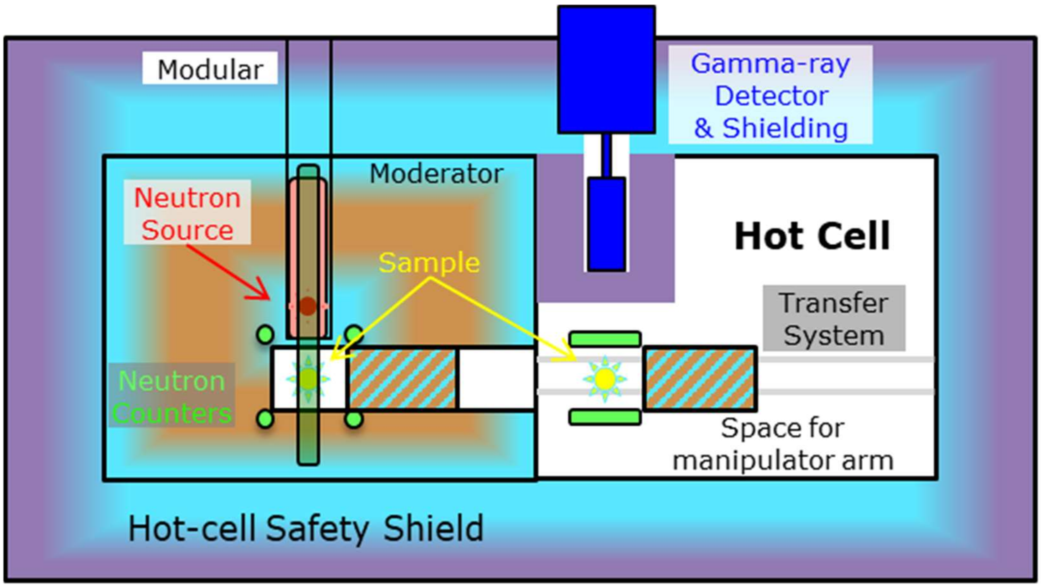


Figure 3. A preliminary final DGS system including neutron detection systems.

Summary

The JAEA/ISCN is developing a compact DGS instrument intended to be used in an analytical laboratory. The JAEA/ISCN is developing neutron detection systems to incorporate into the compact instrument for both neutron source monitoring and fissile mass correlation. Source monitoring will provide a method to normalize the delayed gamma-ray spectra with prompt-fission and delayed-neutron signatures providing a signal to evaluate the fissile mass. To do this, a ^{4}He detector is being investigated for the source monitor since it has low sensitivity to gamma rays and can provide neutron energy information. ^{3}He detectors are intended to be used for neutron signature counters that will provide multiple methods to supplement our delayed gamma-ray mass evaluation. Toward this goal, we are characterizing the neutron detectors and performing model studies to evaluate their integration into the final DGS instrument.

Acknowledgements

This work is supported by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT) under the subsidiary for the "promotion for strengthening nuclear security and the like".

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