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 (3): Analytical Development and Capabilities

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

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

Hee-Jae Lee
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). DGS utilizes neutrons to induce fission in the sample followed by a measurement of gamma rays emitted by the fission products as they decay. The DGS analysis subsequently uses the ratio of the gamma-ray peaks to determine the relative fissile-nuclide contribution for composition evaluation, with integrated counts used to evaluate the fissile mass. This presentation focuses on the analysis being developed to efficiently analyze the gamma rays to evaluate both the composition and the mass to obtain the total fissile nuclide content in the sample. First will be a description of recent results on how the gamma-ray spectrum and the associated analysis is dependent on the interrogation timing. Next, a description will be made regarding the present capability to determine the nuclide content important for safeguards evaluations when combined with the mass determination. Finally, a discussion will be presented on how these experiments affect the final development of the JAEA/IACN DGS Monte Carlo for near-real-time analysis, including present uncertainty quantification.

Keywords: Delayed gamma rays, spectroscopy, analysis, NDA, safeguards, fission, mixed nuclear material

INTRODUCTION

The International Atomic Energy Agency (IAEA) employs safeguards agreements with those states using nuclear power to maintain viable energy capabilities [1, 2, 3]. In particular, the IAEA verifies the state's declared nuclear material amount through defect analysis on the various items, including taking samples [4]. Specifically, the IAEA would like to quantify the total U and Pu content and relative composition found in those materials. The verification of U and Pu nuclides within spent nuclear fuel found throughout the latter half of the nuclear fuel cycle is challenging. Presently, gross item accountancy techniques are used to verify spent nuclear fuel assemblies, which are then sealed and monitored using containment and surveillance techniques [4].

In reprocessing plants, though, the individual assemblies are dismantled and the fuel is dissolved to be processed into purified forms. Consequently, the spent nuclear fuel must undergo partial and bias defect analysis to ensure that significant quantities of the U and Pu are not diverted [5]. To do this, the IAEA performs Hybrid K-Edge Densitometry [6, 4] and Isotope Dilution Mass

Spectrometry (IDMS) [4]. Respectively, these provide the elemental and nuclide content of the spent nuclear fuel solution samples. However, it must be noted that IDMS is a destructive analysis method that requires a time-consuming preparation process that extends the reporting time [7] and produces waste in the lab.

The Japan Atomic Energy Agency Integrated Support Center for Nuclear Nonproliferation and Nuclear Security (JAEA/ISCN) is investigating delayed gamma-ray spectroscopy (DGS) as a method to help improve verification of mixed nuclear materials, especially spent nuclear fuel. This paper will describe the DGS method from the analytical perspective, highlight recent results focused on improving this capability, and the final development needed to make this a viable verification technique.

DELAYED GAMMA-RAY SPECTRSOSCPY METHOD

DGS is an active interrogation technique that uses an external neutron source to induce fission in the dominant U and Pu. This generates fission products mostly with half-lives from a few microseconds to a few hours that decay and emit gamma rays of unique relative intensities. These gamma rays are then observed as peaks within a composite spectrum and used to quantify the sample's composition.

The analysis focuses on evaluating the sample based on known conditions of the detector and the source, which, therefore, must be optimized together with the instrument. For instance, one irradiated fuel assembly emits ~$10^{13\text{-}17}$ gamma rays per second [8,9,10] that requires sufficient filtering and shielding of the detector to reduce count rate. While small samples passively emit significantly fewer gamma rays, the fact that they can be closer to the detector still requires thick filters. To still be able to observe the fission product gamma rays through the filter, the fission rate must be increased by optimizing the neutron flux within the sample [11,12]. The rate can easily be increased by a factor of 500-800 using thermal neutrons by placing moderating material between the source and sample. While this focuses the analysis on the dominant fissile nuclides with large thermal cross-sections (e.g. ^{235}U, ^{239}Pu, and ^{241}Pu), practical sources emit fast neutrons that can also induce fission. Consequently, the analysis requires a highly accurate and precise neutron flux calibration to know from which nuclides the fission products are derived.

The fission products are generated proportionally to both the sample composition and the associated fission yield. Similar to affecting the signal strength, the filter required to suppress the passively emitted gamma rays (e.g. 662-keV from ^{137}Cs) [11,12,10] limits the energy region that can be analyzed to above 2700 keV. Since these high-energy gamma rays must still be quantifiable, a detector with reasonable gamma-ray peak resolution in this energy region is required to observe individual peaks. While high-purity germanium (HPGe) has the best resolution, it is also sensitive to neutron damage and must be kept far from the irradiation source. This source-detector distance will introduce a delay between the irradiation and the measurement, affecting the probability to observe the fission product gamma rays. However, the high-energy gamma rays useful for analysis are largely derived from fission products with half-lives less than 30 minutes and have high production yields. For example, Rb-91 and Tc-106 are important short-lived fission products with half-lives of 58.2 s and 35.6 s since they can be used to differentiate the U and Pu nuclides. On the other hand, Y-95 with a half-life of 618 s is the longest-lived of these and is useful for reference comparison since it is produced at roughly the same yield between the U and Pu nuclides (see Table 1). Consequently, the instrument capabilities and the various fission product half-lives affect the observed spectrum, making the evaluation highly sensitive to the interrogation pattern.

Table 1. Fission products important for DGS analysis.

Fission Product	Half-life (s)	Thermal Fission Yield (%)		
		^{235}U	^{239}Pu	^{241}Pu
Se-86	14.3 (3)	0.84 (2)	0.3 (2)	0.3 (2)
Br-86	55.1 (4)	0.230 (6)	0.2 (1)	0.13 (8)
Br-87	55.68 (12)	1.27 (4)	0.55 (3)	0.44 (7)
Kr-90	32.32 (9)	4.40 (6)	1.10 (3)	1.00 (6)
Rb-90	158 (5)	0.14 (1)	0.14 (9)	0.06 (4)
Rb-90m	258 (4)	0.71 (45)	0.60 (39)	0.3 (2)
Kr-91	8.57 (4)	3.16 (3)	0.70 (1)	0.92 (4)
Rb-91	58.2 (3)	2.22 (3)	1.38 (44)	0.8 (5)
Rb-93	5.84 (2)	3.07 (4)	1.36 (8)	1.5 (5)
Sr-95	23.90 (14)	4.54 (9)	2.61 (118)	2.7 (9)
Y-95	618 (6)	1.11 (35)	1.68 (76)	0.7 (4)
Mo-106	8.73 (12)	0.36 (1)	2.2 (10)	3.9 (12)
Tc-106	35.6 (6)	0.03 (2)	1.83 (11)	1.7 (5)
Te-136	17.63 (8)	1.32 (11)	0.51 (32)	1.7 (5)
I-136	83.4 (1)	1.32 (11)	1.25 (40)	2.0 (6)
Cs-142	1.684 (14)	2.28 (6)	1.4 (3)	2.7 (9)
Ba-142	636 (12)	3.0 (2)	3.1 (5)	1.4 (5)
La-142	5466 (30)	0.10 (3)	0.3 (2)	0.04 (2)

EXPERIMENTS FOR ANALYTICAL IMPROVEMENTS

To evaluate the analytical constraints, the JAEA/ISCN performed experiments in collaboration with the European Commission Joint Research Centre (EC/JRC). Specifically, we tested the gamma-ray spectral sensitivity to the interrogation timing, and, in parallel, tested the instrumentation effects on the spectrum. Early experiments showed that PUNITA in the Ispra (Italy) site of the EC/JRC could be used to obtain delayed gamma-ray spectra with reasonably short delay times [13]. Improving analytical capabilities, an experiment was designed using PUNITA to observe differences in the gamma-ray spectra for timing sensitivity and mass correlation studies. The same U [14] and Pu samples were used as in the earlier studies with five different time patterns that all gave effectively 1-h long total interrogation times [15].

Normalizing to the Y-95 peak count enabled a comparison of peak ratios to be made, independent of neutron source variations and fission rates (see Figure 1). A more accurate peak-ratio was determined by averaging the multiple samples since all dependence on the mass is also removed by using the peak ratios. Toward determining the optimum interrogation pattern to distinguish U and Pu nuclides, the unique gamma rays and the relative differences of those appearing in both spectra were analyzed. Significantly, the U-235 spectrum always had more peaks pass the quality cuts than the Pu-239 spectrum above 2700 keV for all time patterns [16]. Further, the 10-s irradiation/10-s measurement pattern had the largest number of qualifying gamma rays, which decreased with shorter time patterns regardless of the sample. Notably, the 2705-keV peak from Rb-93 is prominent in the U-235 10-s/10-s spectrum due to its 5.84-s half-life, but is overwhelmed by the background in the 60-s/60-s spectrum. Comparatively, the 2701-keV peak from Tc-106 in the Pu-239 spectrum is prominent in all time patterns. Evaluating these two peaks from a mixed-material perspective, they interfere with each other in the 10-s/10-s pattern, but allow the Tc-106 peak to be distinct in the others. Consequently, comparisons with these and other gamma rays resulted in the 60-s/60-s pattern to achieve the best composition determination of these time patterns.

Due to the wide range of U-235 masses in this experiment, a mass correlation study was also performed using the peak counts from each sample [17]. Corrections were made for PUNITA's deuterium-tritium neutron generator temperature variation and the sample's neutron self-shielding and gamma-ray self-attenuation. Integrating above 3300 keV showed a linear correlation with 1.5% differences to the declared masses. Linear correlations were obtained with all of interrogation timing patterns with similar results. Applying the same analysis to the individual peak counts showed a trend, but not a precise correlation.

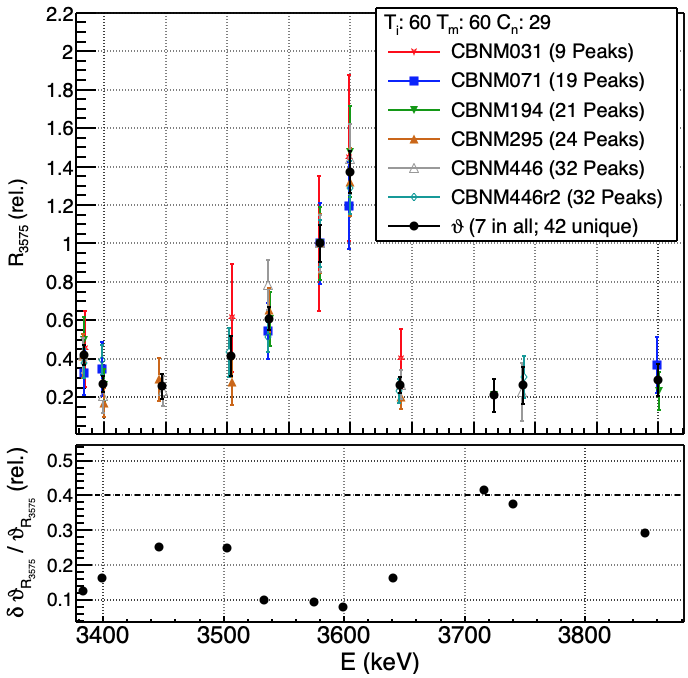

Figure 1. Gamma-ray peak ratios of multiple U-235 sample masses from an irradiation and measurement both of 60 s over 29 cycles [16].

Since the fission product gamma-ray spectrum is also dependent on the instrument itself, similar studies were applied to the results of the JAEA/ISCN Delayed Gamma-ray Test Spectrometer [18]. The DGTS uses a Cf-252 source (DGTS-C), which results in a different moderated neutron flux than that from the D-T generator in PUNITA. Not only does the DGTS Cf-252 emit neutrons with a mean energy lower than a D-T neutron generator, but it also emits them continuously and at a ~20% rate compared to PUNITA. The detector was also connected to a Mirion Technologies LYNX [19] module that allowed us to use multi-spectral scaling (MSS). MSS is an improvement over the pulse-height spectral analysis of PUNITA since enabled a dead-time correction to be applied each second of the measurement periods.

Compared to the short interrogation patterns performed in PUNITA, a 300-s irradiation/300-s measurement pattern was used to highlight the dominant 3575-keV/3600-keV pair of gamma rays. Significantly, the observed spectra had peak intensities similar to PUNITA since the same U and Pu samples were positioned closer to both the source and detector (see Figure 2). Notably, due to the lower mean neutron emission energy, there was no detectable interference in the 2753-keV gamma ray of multiple fission products from Al-27(n,α)Na-24 activation. Additionally, more escape peaks were observed due to the 50% HPGe efficiency compared to the 80% used with PUNITA, showing that the detector choice is important.

Though the DGTS-C and PUNITA experiments focused on different spectral analysis capabilities, both were used to perform U-235 mass correlation studies. After removing the Cf-252 induced activation background, a linear correlation was obtained from DGTS-C spectra with an uncertainty reaching <1% over multiple energy range integrals. Applying this correlation analysis to PUNITA, similar <1% uncertainties were obtained and further evaluations will be made for better quantification. Regardless, these results show that a compact, practical instrument is feasible and can produce reliable DGS signatures.

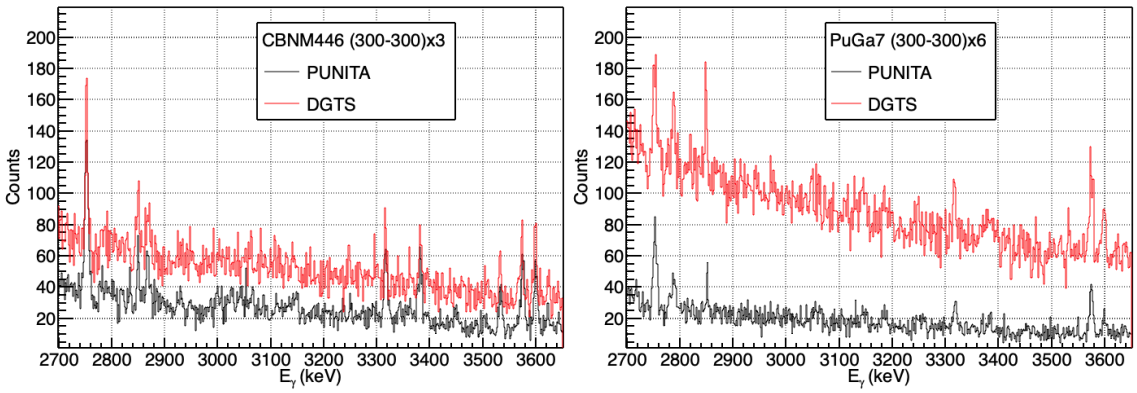

Figure 2. U-235 (left) and Pu-239 (right) spectra comparisons between PUNITA (black) and DGTS (red) interrogations.

ANALYSIS DEVELOPMENT

The JAEA/ISCN is now in the process of verifying the DGS Monte Carlo (DGSMC) [20,21,22] by comparing simulated spectra to these new experimental results. DGSMC and the measured spectra will also be compared to FIER [23] and FISPACT-II [24] to evaluate the different calculation methods. DGSMC is a full Monte Carlo (MC) code that uses the ROOT analysis platform [25] to simulate a DGS interrogation. The relative fission and other reaction rates are determined in the user-defined sample from a flux distribution chosen from MCNP [26]. The subsequent reaction products (e.g. from fission, proton, alpha) are decayed using a hierarchical procedure for each repeated interrogation period (irradiation, delay, measurement, reset). The relative gamma-ray spectrum, including efficiency, is determined from a Geant4 [27] spectrum and spread with an energy-gain calibration from experimental detectors. Comparatively, FISPACT-II was developed using the Bateman equations to calculate the composition of nuclear fuel as it is burned in a reactor, including periods of no flux as if the reactor was off. Though the user can define a single nuclide or mixed material, it was not intended for DGS so the gamma-ray spectrum must be calculated from the decay rate equation. Also uses the Bateman equations, FIER was developed for general fission product decay calculations, like activation experiments, but does not handle the cyclical nature required for a DGS interrogation. However, it evaluates gamma rays emitted over a period of time, like the measurement periods important for DGS, by first calculating the derivative of the Bateman equations.

The validation being performed evaluates the absolute and relative differences between the measured and simulated spectra. Presently, all three codes were used to simulate the PUNITA time-sensitivity experiments for both the U and Pu samples. In all cases, a preliminary analysis showed no significant differences in the short irradiation/short measurement time patterns [28]. DGSMC does show slightly more variation than FIER and FISPACT for the long-measurement patterns, but are still reasonable for comparison. Further, the DGSMC simulated gamma-ray peaks best match the measured gamma-ray peaks of the three codes in the shortest time patterns by using the Geant4 efficiency and escape peaks. Significantly, all three codes show that the 60-s

irradiation, 60-s measurement pattern provides the best distinction between U-235 and Pu-239, matching the measurements.

Final analysis development will be performed to quantify an absolute composition capability with accurate uncertainty quantification. The current composition uncertainty derived by DGSMC inverse MC analysis is sufficient, but can be improved by evaluating those peaks actually observed in the [22]. Once the code provides accurate and precise comparisons, a re-evaluation of the optimum interrogation timing pattern will be performed by scanning all timing patterns possible. Specifically, this will be performed for all spent fuel and other mixed nuclear material sample compositions, dominantly based on the spent fuel library examples [29]. Additionally, this will be applied to different form factors, from reprocessing plant input solutions to full assemblies. Factoring in the conditions for small sample evaluations will verify the expected 10^9 neutron-per-second source emission rate required for spent fuel solutions [11,12]. From this, analytical capabilities will be established for refining the DGS instrument design, focusing on the present small sample instrumentation, then expanding into other form factors.

SUMMARY

The JAEA/ISCN has been performing experiments with the EC/JRC in order to develop DGS for nuclear safeguards capabilities. The primary requirement for DGS is to develop a viable analysis to accurately and precisely determine the fissile nuclide composition in mixed nuclear material samples. The JAEA/ISCN is focusing this DGS work on spent nuclear fuel, which is constrained by the passive emissions and material form factor. Recent experiments using PUNITA show that the DG from short-lived fission products have clear distinction between U-235 and Pu-239 samples as well as a linear mass correlation. These were confirmed using the JAEA/ISCN designed DGTS using Cf-252 as the external source, both experiments showing <1% mean uncertainty in the mass determination. These measured spectra are being compared to the JAEA/ISCN DGSMC and two other production/decay calculation codes, which all show similar results to the measurements. Further improvements are underway to expand the reliability and capability of DGSMC for safeguards verification applications.

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”. The authors would like to thank Bent Pedersen, Kamel Abbas, and their teams in the EC/JRC for the experimental data collected under the agreement between JAEA and EURATOM in the field of nuclear material safeguards research and development.

REFERENCES

[1] United Nations, “Treaty on the Non-Proliferation of Nuclear Weapons (NPT)”, https://www.un.org/disarmament/wmd/nuclear/npt/text, last accessed May 11, 2020.

[2] International Atomic Energy Agency, “The structure and content of agreements between the agency and stated required in connection with the treaty on the non-proliferation of nuclear weapons”, INFCIRC/153, 1972.

[3] International Atomic Energy Agency, “Model protocol additional to the agreement(s) between state(s) and the International Atomic Energy Agency for the application of safeguards”, INFCIRC/540, 1997.

[4] International Atomic Energy Agency, “Safeguards Techniques and Equipment: 2011 Edition”, Int. Nucl. Verif. Series No. 1, Rev. 2, 2011.

[5] S.J. Johnson et al., "Development of the Safeguards Approach for the Rokkasho Reprocessing Plant", IAEA-SM-367/8/01, 2001.
[6] H. Ottmar, H. Eberle, "The Hybrid K-Edge/K-XRF Densitometer: Principles – Design – Performance", KfK 4590, 1991.
[7] T. Itoh et al., "Enhanced Cooperation between SSAC and IAEA through Joint Operation of On Site Laboratory for Safeguarding Rokkasho Reprocessing Plant", IAEA-CN-184/70, 2010.
[8] D.C. Rodriguez, "Ultra-High Rate Measurements of Spent Fuel Gamma-ray Emissions", APS April Meeting, 2014.
[9] J.E. Fast, J.W. Chenault, B.D. Glasgow, D.C. Rodriguez, "Spent Nuclear Fuel Measurements", PNNL-23561, Pacific Northwest Nat. Lab., 2014.
[10] L.W. Campbell, L.E. Smith, A.C. Misner, "High-Energy Delayed Gamma Spectroscopy for Spent Nuclear Fuel Assay", IEEE Trans. on Nuc. Science 58 (1), 2011, 231-240.
[11] D.C. Rodriguez, F. Rossi, T. Takahashi et al., "Model design of a compact delayed gamma-ray moderator system using ^{252}Cf for safeguards verification measurements", Applied Rad. and Isotopes 148, 2019, 114-125.
[12] F. Rossi, M. Koizumi, D.C. Rodriguez, "Model design of a deuterium-deuterium neutron generator moderator and evaluation for delayed gamma-ray nondestructive assay for safeguards verification", J. of Nuc. Science and Tech. 58 (3), 2021, 302-314.
[13] D.C. Rodriguez, M. Koizumi. F. Rossi et al., "Utilizing PUNITA experiments to evaluate fundamental delayed gamma-ray spectroscopy interrogation requirements for nuclear safeguards", J. of Nuc. Science and Tech. 57 (8), 2020, 975-988.
[14] de Bievre P, Eschbach HL, Lesser R et al., "^{235}Uranium isotope abundance certified reference material for gamma spectrometry EC nuclear reference material 171 certification report," European Commission Joint Research Centre, Geel, Belgium, Tech. Report EUR-10503-EN, 1986.
[15] D.C. Rodriguez, M. Koizumi, H.J. Lee et al., "JAEA-JRC Collaborative development of Delayed Gamma-ray Spectroscopy for Nuclear Material Evaluation (1): Project Overview", 61st INMM Annual Meeting, 2020.
[16] D.C. Rodriguez, T. Bogucarska, M. Koizumi et al., "Evaluation of high-energy delayed gamma-ray spectra dependence on interrogation timing patterns", Nuc. Inst. and Methods A 997, 2021, 165146.
[17] F. Rossi, T. Bogucarska, M. Koizumi et al., "Correlating the fissile mass of standard uranium samples with delayed gamma rays from fission products", Nuc. Inst. and Methods A 977, 2020, 164306.
[18] D.C. Rodriguez, K. Abbas, M. Koizumi et al., "Development and testing of a Delayed Gamma-ray Spectroscopy instrument utilizing Cf-252 neutrons evaluated for nuclear safeguards applications", Nuc. Inst. and Methods A, Accepted July 2021.
[19] Mirion Technologies, "Radioactivity Measurement Equipment, Edition 2.0", 2020.
[20] Rodriguez DC, Rossi F, Seya M, Koizumi M, "Delayed gamma-ray spectroscopy inverse Monte Carlo analysis method for nuclear safeguards nondestructive assay applications", IEEE Nuc. Sci. Symposium, 2017.
[21] Rodriguez DC, Rossi F, Takahashi T et al., "Development of Delayed Gamma-ray Spectroscopy for Nuclear Material", INMM 59th Annual Meeting, 2018.
[22] D.C. Rodriguez, K. Abbas, J.M. Crochemore et al., "Utilizing nuclear data in delayed gamma-ray spectroscopy inverse Monte Carlo analysis", International Conference on Nuclear Data for Science and Technology, 2019, EPJ Web Conf. 239, 17005.
[23] E.F. Matthews, B.L. Goldblum, L.A. Bernstein et al., "FIER: Software for analytical modeling of delayed gamma-ray spectra", Nuc. Inst. and Methods A 891, 2018, 111-117.

[24] J.Ch. Sublet, J.W. Eastwood, J.G. Morgan et al., “FISPACT-II: An Advanced Simulation System for Activation, Transmutation and Material Modelling”, Nuc. Data Sheets 139, 2017, 77-137.
[25] R. Brun, F. Rademakers, “ROOT: An Object-Oriented Data Analysis Framework”, Nuc. Inst. and Methods A 389, 1997, 81-86.
[26] T. Goorley et al., “Initial MCNP6 Release Overview”, Nuc. Technology 180, 2012, 298-315.
[27] S. Agostinelli, J. Allison, K. Amako et al., “Geant4 – a simulation toolkit”, Nuc. Inst. and Methods A 506 (3), 2003, 250-303.
[28] Workshop on Nuclear Data for Reactor Antineutrino Measurements, June 21-24, 2021.
[29] H.R. Trellue, M.L. Fensin, J.R. Richard et al., “Description of the Spent Nuclear Fuel Used in the Next Generation Safeguards Initiative to Determine Plutonium Mass in Spent Fuel”, LA-UR-11-00300, Los Alamos National Laboratory, 2010.